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Executive Summary

The option of transuranic recycling in U. S. LWRs has the advantage of exploiting a large capacity of nuclear reactor systems and making progress in the near-term by demonstrating the benefits of a nuclear fuel cycle which incorporates spent fuel reprocessing and recycling. Experience has already been gained in European (particularly, France) and Japanese nuclear programs from utilizing MOX fuel fabricated with reactor-grade plutonium, separated from spent uranium and the other transuranics using the PUREX process. While employing this approach recovers valuable energy (from plutonium fission) which would be discarded in a once-through UO_2 fuel cycle, the issue of Am-241 disposition, which contributes to repository heating and essentially limits the repository loading, would remain.

In the present study, the effectiveness of limited homogeneous recycling of plutonium, neptunium, and americium in LWRs for improving the repository performance relative to a once-through fuel cycle has been investigated. The recycled transuranics were loaded into fuel pins in MOX, CORAIL (heterogeneous MOX and UO_2), and inert-matrix (IMF) fuel assemblies. Studies of the viability of using heterogeneous targets for americium and curium transmutation are also being performed within the AFCI program. Charge and discharge isotopic vectors and mass flows were determined, and the consequent impacts on safety parameters and repository heat load were estimated.

The basic assumptions employed in this study included:

- High-burnup UO_2 (51 or 45 GWd/MTHM)
- Minimal post-irradiation cooling time (5 years) before spent fuel reprocessing and TRU recycling
- Separated cesium, strontium, and depleted uranium sent to interim storage
- Separated curium assumed sent to repository for permanent disposal
- Direct disposal of spent fuel assemblies from the final recycle stage

For the MOX and IMF recycling strategies, the Pu+Np+Am in the charged assembly in recycle N was derived solely from discharged assemblies in recycle N-1. Both strategies exhibited a rather rapid burndown of the fissile plutonium. Consequently, the transuranic loading needed to maintain the reactor cycle length in successive recycles had to be increased beyond what is practical from the viewpoint of safety coefficients fuel handling parameters, but this approach is the most straightforward and predicts the maximum heat load benefit achievable for a given number of recycles. For the IMF strategy in particular, nothing more than a single recycle is considered feasible unless the recycled fuel is supported with reactor-grade material (i.e., that derived from spent UO_2) or low-enriched uranium. Future studies which refine the recycling strategies to include blending schemes or heterogeneous IMF/UO_2 assembly designs should be considered.

Because of the heterogeneous design of the CORAIL assembly, it is most practical to co-process the spent UO_2 and MOX pins; distinguishing and segregating spent UO_2 and MOX pins would be difficult. Thus, the CORAIL assembly concept incorporates a recycling strategy which blends reactor-grade and recycled TRU. Utilizing this approach, the material can be multi-recycled without encountering operations constraints. As presently formulated this approach slowly stabilizes the inventory of transuranics, but does not burn down the TRU in the existing stockpile.

Pu+Np+Am recycling in homogeneous MOX assemblies provides the smallest repository benefit for a fixed number of recycles, although this recycling strategy is the most straightforward approach given current experience in Europe and Japan. A single recycle in this fuel form yields a 9% increase in the effective loading of spent fuel in the repository based on thermal limits in the current "high-temperature operating mode." Continued recycling through up to 5 recycles of the Pu+Np+Am was considered, resulting in a repository loading increase of 49% relative to the once-through fuel cycle. However, the large transuranic loading at this point (>50% TRU/HM) presents challenges for fuel fabrication (materials), reactor operations (safety coefficients), and fuel handling (dose to workers).

The CORAIL concept provides a modest gain on the repository loading relative to the homogeneous MOX approach. If only a single recycle is practiced, the repository capacity can be theoretically increased by 20%, while if 5 recycles are practiced, there is about a 70% improvement relative to the once-through fuel cycle. The utilization of an inert-matrix fuel form such as solid solution $(TRU,Zr)O_2$ yields the greatest benefit because no additional transuranics are produced during the fuel irradiation. A single recycle provides a loading increase of 80%, but further recycles, although theoretically possible, are not considered practical without enhancing the recycling strategy.

Recycling plutonium and americium in MOX, CORAIL, and IMF fuel forms contributes directly to the repository loading benefit by reducing the masses of Am-241 chain nuclides (Pu-241+Am-241) disposed in nuclear waste, and their associated decay heat, relative to the once-through fuel cycle. Neptunium was also recycled in these scenarios because of the potential reduction of the released dose from the repository, although the dose reduction was not quantified in this study. However, recycling the neptunium penalizes the loading benefit somewhat due to the increased disposal of Pu-238 and its associated decay heat. An assessment of the trade-offs between dose reduction and increased decay heat due to neptunium recycling could be performed. The curium extracted from spent UO_2 was not recycled in the LWR recycling strategies evaluated here because of its added complications to fuel handling.

It was found that the longer reprocessing and recycling of the material in the spent UO_2 is delayed, the less is the achievable benefit. The consequences of delaying recycling are most severe for the IMF fuel form. Ongoing decay of fissile Pu-241 to Am-241 increases the required TRU loading, hardening the neutron spectrum in a way that negatively impacts the Pu-241 and Am-241 transmutation rates. Extending the cooling time from 5 to 20 years from reactor discharge to recycling of the Pu+Np+Am in IMF, the repository loading benefit is reduced by a factor of 4, or a 20% loading increase relative to a once-through fuel cycle.

Lastly, the impact of the transuranic separations strategy employed for actinide management was investigated by comparing the results obtained here with those obtained from earlier studies. In particular, the discharged actinide masses (normalized to the net energy produced by the transuranics and their source UO_2) after a single LWR recycle were compared for Pu, Pu+Np, Pu+Np+Am, and all TRU separations strategies. Mono-recycling of separated plutonium burns fissile Pu-239 and would lower the attractiveness of the repository as a "plutonium mine." However, the normalized mass of Am-241 and Np-237 chain nuclides are actually increased by this approach relative to a once-through

fuel cycle. The implication is that this elevates the long-term decay heat which limits the waste loading in the current repository design.

Recycling the americium appears necessary for an actinide management strategy that benefits the repository in terms of reduced heat load and reduced released dose, although the gains which can be realized from limited recycling in an LWR are considered marginal in light of a continued utilization of nuclear power and generation of spent fuel. Whether nuclear power grows or even continues at its current capacity, the gains realized here are not large enough to stave off the need for a second repository unless further recycling in LWRs or future, advanced systems is practiced. Nonetheless, actinide management in LWRs is sensible, particularly as a demonstration of spent fuel reprocessing and recycling technologies to be employed in Generation-IV systems and/or as a technique for reducing the hazardous components in legacy spent nuclear fuel.

However, challenges to transuranic recycling in current-generation LWR systems do exist and these must be considered either as topics for future technical work or as factors in making decisions regarding the viability of employing LWRs for actinide management. There is, first of all, the issue of fuels development and utilizing fuel forms which have not been commercially fabricated or extensively tested. MOX fuel is being used in international nuclear programs, but this has been limited to separated plutonium; inert-matrix fuels are just now being considered, and questions about reprocessing on the back-end of the fuel cycle must be addressed. Second, there are the concerns over the proliferation-resistance of utilizing a fuel cycle based on separation of transuranics from spent nuclear fuel. There does not appear to be a single technical argument to address all concerns, but, rather, intrinsic (e.g., isotopics) and extrinsic (e.g., security) safeguards will be necessary. Third, there remains the fundamental question of whether U. S. nuclear utilities will consider recycled fuel to be an attractive resource. At the heart of this issue are reliability (how will the types of fuels considered affect day-to-day reactor operations), reactor safety (are current safety systems designed for UO_2 fuel adequate), and economics (what will be the ultimate cost or benefit to the utility).

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HOMOGENEOUS RECYCLING STRATEGIES IN LWRS FOR PLUTONIUM, NEPTUNIUM, AND AMERICIUM MANAGEMENT

Abstract

Strategies for actinide management which employ homogeneous recycling of plutonium, neptunium, and americium are investigated. The benefits of the actinide management strategy are measured by gains in the temperature-constrained repository loading. Because curium contributes little to the repository heat load, and because recycling curium complicates the LWR fuel cycle, it is presumed to be separated from spent UO_2. A number of options are available for dealing with the curium stream, including permanent disposal, transmutation in heterogeneous targets, or future recycling in fast-spectrum systems.

Mixed-oxide (MOX), CORAIL (a heterogeneous MOX and UO_2 fuel assembly design), and inert-matrix (IMF) fuel forms for LWR-based transmutation are considered. It is anticipated that the greatest benefit will be realized from the utilization of IMF, but this approach requires development of fuel fabrication and spent fuel reprocessing technologies. The impact of delaying spent UO_2 reprocessing and recycling in these fuel forms is also evaluated.

The impact of the transuranic separations strategy is also investigated by comparing the results obtained here with those from previous actinide management studies involving plutonium, plutonium plus neptunium, and all transuranic recycling.

I. INTRODUCTION

Various recycle strategies have been proposed to manage the inventory of transuranics in commercial spent nuclear fuel (CSNF), with a particular goal of increasing the loading capacity of spent fuel and reprocessing wastes in the Yucca Mountain repository. Transuranic recycling in commercial LWRs can be seen as a viable means of slowing the accumulation of transuranics in the nationwide CSNF stockpile. Furthermore, this type of approach is an important first step in demonstrating the benefits of a nuclear fuel cycle which incorporates recycling, such as envisioned for Generation-IV reactor systems under development. Recycling strategies of this sort are not proposed as an attempt to eliminate the need of a geologic nuclear waste repository, but as a means to enhance the usefulness of the repository currently under construction in the U. S., perhaps circumventing the need for a second facility. A US-DOE Secretarial recommendation on the need for the construction of a second geologic repository is required by 2010.

On the other hand, challenges to transuranic recycling in current-generation LWR systems do exist and these must be considered as factors in making decisions regarding the viability of employing LWRs for actinide management. In general terms, the issues to be considered are fuels development, proliferation resistance, and attractiveness of recycled fuel to U. S. nuclear utilities. These issues are outside the scope of the current study, but are noted as topics for future study.

Although reductions of the long-term radiotoxicity of the disposed material and the dose released from the repository are important goals, they are not deemed to be active constraints on the repository capacity in its current design. Rather, a constraint on the temperature of the "rock pillar" in the mountain (the area midway between the storage tunnels, or drifts) appears to limit the loading of directly disposed, once-through spent fuel in the current "high-temperature operating mode." A methodology to perform detailed time-dependent thermal analyses of the repository with different waste package characteristics has been developed by analysts in the ANL Fuel Cycle Modeling section. This methodology has been used to assess the impact of proposed recycling strategies on the loading capacity of the repository.

It has been repeatedly shown that for a once-through fuel cycle with direct disposal of the spent fuel assemblies, the major contributors to the limiting temperature of the repository rock pillar are a few key isotopes of plutonium and americium. Previous studies have shown benefits ranging from factors of 3 to 50 increase in repository capacity with *multi-recycling* of plutonium and americium (thus limiting their disposal to processing wastes only) and select fission product removal.

However, even with anticipated license extensions, the current generation of LWRs used for commercial power production in the United States will begin shutting down in 2010, and will likely be completely retired by 2050.[1] The next generation of nuclear power reactors will be more favorably designed for transuranic utilization, driven by the natural accumulation of plutonium stockpiles in spent uranium-based fuels, as well as possible limitations in uranium ore reserves. Thus, the types of LWR recycling campaigns which have been considered will be feasible for no more than 30 years, assuming an aggressive program of fuels development and infrastructure construction (e.g. spent fuel reprocessing and advanced fuel fabrication facilities).

Thus, the focus of this report is an assessment of the potential benefit of a *finite number of recycles* of transuranics in an LWR, with disposal of the material not consumed at the end of the recycling campaign. The temperature-constrained repository loading is used as a measure for comparing the performance of various recycling options which are distinguished by fuel form, cooling time before reprocessing, and the transuranic species being recycled. This report deals with so-called "homogeneous recycling" approaches, in which the transuranic material is fabricated into power-producing fuel pins containing a mixture of fissile (e.g., Pu-239) and non-fissile (e.g., Am-241) nuclides. Heterogeneous recycling options, in which americium and curium (which are dominated by non-fissile isotopes) are fabricated into target pins for transmutation in an LWR, are being investigated in a separate study being performed at the Oak Ridge National Laboratory.

The Advanced Fuel Cycle Initiative (AFCI) has supported a breadth of work to evaluate the ideal transuranic separation and recycle strategy. Previous studies of LWR-based transmutation performed at ANL have considered the benefits of homogeneously recycling plutonium, plutonium and neptunium, and all transuranic species.[2,3,4,5] A study of a wide range of hypothetical separation schemes (Pu, Pu+Np, Pu+Np+Am, etc.) with multi-recycling has also been performed, focusing on the proliferation resistance of the various fuel cycles and fuel handling issues.[6]

The primary separation strategy considered in this study is plutonium, neptunium, and americium recycling. Plutonium and americium are recycled in order to reduce the intermediate term (100 to 1500 years after spent fuel irradiation) decay heat of the disposed waste which accounts for the bulk of the repository heating. Since the long-term released dose from the repository is dominated by neptunium, it is sensible to consume it by transmutation in a reactor, as well. Furthermore, neptunium is easily separated with plutonium in aqueous reprocessing techniques. The actinide masses remaining after a single recycle in an LWR are cross-compared with results from other separation hypotheses considered in previous studies.

Curium accounts for ~0.6% of the TRU mass in spent UO_2 (~0.008% of the heavy metal). The curium was not considered to be homogeneously recycled in this study because of the high heat and radiation source characteristics of Cm-244. As an alternative to permanent disposal in the repository, the curium could be fabricated into dedicated targets for LWR transmutation, recycled in a fast spectrum system along with transuranics remaining after the LWR recycling campaign, or placed in interim storage to allow for cooling/decay to Pu-240 before recycling in either an LWR or fast system. It should be noted, however, that handling the curium in these alternative options is not simple. Analyses of curium storage options have revealed that decay heat (product temperature) and He generation from α-decay (accumulating gas pressure) are crucial considerations in developing a suitable engineered storage form.[7]

Section II of this report reviews the benefits and consequences of various separation strategies for transuranics management. Evaluations of the approaches considered in this study for transuranic management are discussed in Section III. The performance measures (temperature-constrained repository loading) of the various homogeneous recycling scenarios are compared in Section IV. This section includes an analysis of the impact of fuel form and cooling time on the material masses discharged from the recycling scenario. Lastly, the conclusions of the study are given in Section V.

II. TRANSURANIC SEPARATION STRATEGIES

Nuclear waste "packages" will be placed end-to-end in underground tunnels at the Yucca Mountain site. Forced-air cooling of the repository effectively removes the decay heat generated by the waste packages, so that the rock pillar temperature is largely controlled by the integrated decay heat generated from the time the cooling fans are turned off to the time that the rock reaches its peak temperature. It is reasonable to assume that the fans will be turned off 100 years after the spent fuel (or consequent waste material) disposed in the repository was originally discharged from the reactor. Without transuranic recycling, spent UO_2 fuel assemblies will be placed in waste packages and directly disposed in the repository.

If 90% or more of the plutonium and americium are removed from the spent fuel before the waste material is permanently disposed in the repository, detailed thermal analyses have shown that the current temperature constraints on the repository can be met with a nuclear waste loading equivalent to 4-5 times that for direct disposal of spent UO_2.[8] The mechanism for eliminating the plutonium and americium from permanently disposed waste is largely irrelevant (i.e., transmutation in thermal- and/or fast-spectrum systems can be employed), although certain systems or fuel forms may present advantages over others based on physics, economics, or technology readiness considerations. If, additionally, cesium and strontium are removed and placed in interim storage until they are sufficiently cooled (the half-lives of the dominant Cs-137 and Sr-90 are around 30 years), the same analysis has shown that the effective repository loading can be potentially increased by a factor of 43.

In this instance, the loading is constrained by the imposed 200°C limit for the temperature of the drift wall, which occurs at the time of waste emplacement. The dominant source of the decay heat in this case (with Pu, Am, Cs, and Sr removed) is Cm-244. With this in mind, the logical next step to increasing the usefulness of the Yucca Mountain site would be transmutation of the curium in the spent fuel. It has been reported in technical presentations that based only on thermal constraints, limiting the disposal of Pu, Np, Am, Cm, Cs, and Sr to reprocessing wastes could enable a factor of 100 increase in the repository loading relative to disposal of once-through UO_2.[9] The practicality of accomplishing this by employing LWRs alone must be taken into consideration.

Table 1 summarizes certain radio-isotopic properties for an initial unit mass of selected actinides. The data provided here can be used to provide a preliminary estimate of the impact these nuclides could have on waste disposal (integrated decay heat, radiotoxicity) and handling issues for recycled fuel (instantaneous decay heat, neutron and γ radiation sources). These properties were calculated using the ORIGEN2[10] code, and for the integrated decay heat and radiotoxicity data include the contributions of the parent nuclide and all daughter nuclides generated during the time-frame of interest. For reference purposes, the mass of each nuclide in 1 metric ton of spent UO_2 is included in Table 1.

The integrated decay heat data/unit mass provides a measure of which actinides should be limited in the disposed waste if the rock pillar temperature is an active constraint on repository loading. For direct disposal of spent UO_2 at its maximal loading,

Table 1. Radio-Isotopic Properties for an Initial One Gram of Selected Actinides.

	Integrated Decay Heat, 100-1250 Years (Watt-years)	Radiotoxicity[1] at 10,000 Years (Sv)	Radiotoxicity at 200,000 Years (Sv)	Instantaneous Decay Heat (Watts)	*Neutron Source (neutrons/s)*		γ Source (MeV/s)	Mass in Spent UO_2[3] (g/MTIHM)
					(α,n)[2]	Spontaneous Fission		
U-235	7.4E-05	0.03	0.13	5.7E-8	7.91E-4	3.53E-4	1.24E+4	7,684
U-238	1.2E-05	6.8E-4	9.1E-3	8.5E-9	1.06E-4	0.013	1.47E+1	921,010
Np-237	0.03	2.68	11.60	2.2E-5	0.50	0.0	8.80E+5	618
Pu-238	33.07	51.06	360.05	0.568	16,290	2,657	1.01E+9	307
Pu-239	2.16	327.70	1.50	0.002	45.30	0.023	1.44E+6	6,185
Pu-240	7.60	554.99	0.11	0.007	172.7	910.5	1.29E+7	2,926
Pu-241	52.87	2.61	11.39	0.003	0.0	0.0	1.45E+7	1,377
Am-241	51.13	2.62	11.39	0.114	3,293	1.24	3.10E+9	439
Am-243	7.57	687.15	2.07	0.006	170.1	3.35	4.07E+8	196
Cm-244	32.68	547.30	0.11	2.831	92,360	11,120,000	4.58E+9	69

[1]Cancer dose from ingestion.
[2]Calculated by ORIGEN2 assuming material in an oxide matrix.
[3]4.3 wt.% U-235, 51 GWd/MTIHM burnup, 5 years post-irradiation cooling.

the rock pillar reaches its peak temperature of 96°C about 1,500 years after irradiation of the spent fuel. Allowing time for heat transport from the waste package into the region between the drift tunnels, it appears that the decay heat integrated from 100 to 1,250 years after irradiation will provide the best measure of the contributors to the limiting temperature. The values in Table 1 were determined by numerically integrating decay heat data calculated by ORIGEN2 over the specified time interval,

$$I_j = \sum_i \sum_l \frac{H_{i,t_l} + H_{i,t_{l+1}}}{2}(t_{l+1} - t_l), \quad (1)$$

where the summation over i includes all daughter nuclides of nuclide j and $H_{i,t}$ is the decay heat of nuclide i at time t_l. The time step length (t_{l+1}-t_l) in the interval of integration ranged from 25 to 250 years.

The concentrations of some of these actinides (and their daughters) in the disposed waste will be naturally limited because of their relative concentration in the spent fuel, as indicated in the last column in Table 1. Even so, a unit mass of Pu-241 contributes the most to the temperature of the rock pillar, highlighting that this is a primary candidate for recycling and transmutation. The heating associated with Pu-241 actually comes from its Am-241 daughter, so Am-241 is also typically targeted for transmutation. Following the Am-241 chain, limiting Pu-238 in the disposed waste appears next in order of importance. The relatively high integrated decay heat value for Cm-244 indicates the potential difficulty of permanently disposing separated curium in a concentrated form, although the true difficulty in this case probably lies in the heat at the time of disposal, as discussed above.

The radiotoxicity values reported in Table 1 are the cancer dose from ingestion of one gram of the parent nuclide and all its daughters following 10,000 or 200,000 years of decay, and were calculated using dose commitment factors derived from ICRP Publication 60.[11] The current regulatory period for Yucca Mountain is 10,000 years, and performance modeling for the repository reported in Ref. 12 shows that the waste package containers do not fail during this period. Thus, while Am-243 (and other actinides) has a high value for specific radiotoxicity at 10,000 years, it is unlikely that it or any of its daughters will be released from the repository within this time period.

The specific radiotoxicity values calculated at 200,000 years are considered more important for estimating the contributors to the released dose from the repository. Limiting the disposal of Np-237 and/or its parents Am-241 and Pu-241 is considered the most crucial for reducing the magnitude of the long-term released dose based on performance modeling results reported in Ref. 12. It is notable that the specific radiotoxicity for Pu-238 is significantly higher, however. The cancer dose in this case does not come from the Pu-238 itself, but from its daughters Po-210, Pb-210, Ra-226, and Th-230 (202, 91, 35, and 25 Sv/g Pu-238, respectively). For some nuclides, such as Po-210, multiple dose commitment factors are available in Ref. 11 based on the chemical form in which the nuclide is present in the body. The higher values were used to calculate the radiotoxicity values in Table 1, which may be an overly conservative approach.

As stated above, if the disposal of all transuranic nuclides were limited to reprocessing losses, the current temperature constraints on the repository could be met with a nuclear waste loading equivalent to 100 times that for direct-disposed spent UO_2. Multi-recycling all transuranics in an LWR has been shown to be feasible from a neutronics viewpoint using the CORAIL fuel cycle concept.[4] However, an LWR recycle approach to transuranic management does present challenges, particularly in regard to fuel handling.

The instantaneous radio-isotopic properties provided in Table 1 give a measure of the fuel handling impact at the fabrication plant or reactor site when particular actinide species are recycled. The heat and radiation sources for the uranium isotopes are quite low compared to other actinides, and for this reason fuel handling procedures developed for current U. S. nuclear power plants allow for contact handling of the fabricated UO_2 assemblies, as well as lower shielding requirements than required for mixed-oxide (MOX) fuel.

It was reported in Ref. 5 that the decay heat for a typical MOX assembly fabricated with plutonium is around 800 watts (compared with 0.03 W for a UO_2 assembly), and that most of this heat comes from Pu-238. Thus, it is desirable to limit the assembly loading of Pu-238, particularly if special post-fabrication assembly storage is to be avoided. According to the data presented in Table 1, recycling Cm-244 presents an even greater challenge. If the above MOX assembly included just 250 grams Cm-244 (a concentration of ~0.7% of the TRU mass), the fabricated assembly would produce the same heat as a spent UO_2 assembly after 5 years cooling (~1.5 kW).

Radiation dose to fuel handling workers is a greater consideration for transuranic-bearing fuels. Photon dose rates from unirradiated MOX fuel assemblies fabricated with plutonium are significantly higher than those from UO_2, but are manageable without altering fuel handling equipment at the reactor site. Indeed, the fuel cladding provides significant shielding against the lower-energy photons emitted by Pu-238 and, to a lesser extent, those emitted by Am-241. Because of the higher photon energy source from Am-241, it can be expected that a limit will be placed on the Am-241 concentration in the MOX material at the fabrication facility. For example, the Am-241 concentration at the French MELOX plant is limited to <3% Am-241/Pu to prevent excess radiation exposure to workers.

The photon source/gram is also high for Cm-244, but as for Pu-238, these are very low energy photons which are relatively easy to shield. A much larger dose contribution results from the extremely high neutron source from spontaneous fission, which is more difficult to shield against. Furthermore, if the curium is multi-recycled in the LWR, the buildup of very high mass actinides (i.e., Cf-252) in even minute quantities results in a neutron source rate that is more than four orders of magnitude larger than that for conventional MOX fabricated from plutonium only; calculations performed in Ref. 6 found that the emitted dose rate from the fuel pin is increased by a factor of 11,000.

For these reasons, it is envisioned that transuranic recycling in current-generation LWRs will be limited to plutonium, neptunium, and americium, and the relative concentrations of these elements in the fuel must be controlled to limit the radiation shielding requirements for workers at a fabrication facility. An identification of these

limits could be the focus of future work. Upgrading fuel handling equipment and/or practices at current reactor sites to accommodate curium recycling in LWRs would likely be prohibitively expensive.

Table 2 summarizes the integrated decay heat contribution of heavy metal nuclides in spent UO_2 irradiated to a discharge burnup of 51 GWd/MT. Before integration, the decay heat was normalized to the thermal energy produced by the fuel during its residence in the reactor. Thus, the key actinide contributors to the rock pillar temperature in a once-through fuel cycle scenario are Am-241, Pu-238, Pu-239, and Pu-240. It is noted that the contribution to the integrated decay heat from all fission products ranges from 7% to less than 4%, depending on the time interval of integration. If spent fuel processing is utilized and the cesium and strontium (and their short-lived daughters) are eliminated from the permanently disposed material, the fission product contribution is practically negligible

It is important to note that while roughly 65% of the relevant integrated decay heat is generated by Am-241, only about one-fourth of the Am-241 in the spent UO_2 at 100 years is present at 5-years post-irradiation cooling (the earliest time at which spent fuel processing is practically considered); the major contributor to the Am-241 in the repository is the *in situ* decay of Pu-241. Thus, the primary target of most LWR transmutation strategies is the destruction of Pu-241 (as well as Am-241) in advanced fuel forms, such as mixed-oxide (MOX) or inert-matrix fuels (IMF).

Neptunium was also considered to be recycled in this study. This choice is driven partly by the ease with which neptunium separates with plutonium from spent nuclear fuel. Also, the long-term released dose from a Yucca Mountain-type repository is dominated by Np-237, so it is sensible to consume it by transmutation in a reactor as long as its presence in the recycled fuel does not severely penalize other performance parameters (e.g., reactor cycle length). It should be kept in mind, however, that only 25% of the Np-237 inventory 200,000 years after disposal of spent UO_2 (the approximate time at which the calculated released dose reaches its peak) is present at discharge. The remaining 75% comes from the *in situ* decay of Pu-241 and Am-241, which again highlights the benefit of plutonium and americium recycling.

Table 2. Contributors to Direct-Disposed UO_2 Integrated Decay Heat (Watt-years/GWd).

Nuclide	80-1000 years		100-1000 years		100-1250 years		100-1500 years		100-1750 years	
U-234	1.50	0.06%	1.48	0.06%	1.91	0.07%	2.33	0.08%	2.76	0.08%
U-235	0.01	0.00%	0.01	0.00%	0.01	0.00%	0.01	0.00%	0.01	0.00%
U-236	0.19	0.01%	0.18	0.01%	0.23	0.01%	0.29	0.01%	0.34	0.01%
U-238	0.14	0.01%	0.14	0.01%	0.18	0.01%	0.22	0.01%	0.25	0.01%
Np-237	0.61	0.02%	0.60	0.02%	0.82	0.03%	1.05	0.03%	1.29	0.04%
Np-239	1.64	0.06%	1.61	0.06%	2.03	0.07%	2.44	0.08%	2.84	0.09%
Pu-238	**241.24**	**9.17%**	**206.27**	**8.23%**	**206.47**	**7.27%**	**206.51**	**6.64%**	**206.51**	**6.18%**
Pu-239	**209.79**	**7.98%**	**205.17**	**8.19%**	**261.32**	**9.20%**	**317.11**	**10.19%**	**372.52**	**11.15%**
Pu-240	**361.76**	**13.76%**	**353.52**	**14.10%**	**445.88**	**15.70%**	**535.83**	**17.22%**	**623.43**	**18.66%**
Pu-241	0.05	0.00%	0.02	0.00%	0.02	0.00%	0.02	0.00%	0.02	0.00%
Pu-242	1.76	0.07%	1.72	0.07%	2.20	0.08%	2.67	0.09%	3.15	0.09%
Am-241	**1781.71**	**67.75%**	**1709.93**	**68.22%**	**1886.86**	**66.45%**	**2005.39**	**64.46%**	**2084.80**	**62.40%**
Am-242m	0.01	0.00%	0.01	0.00%	0.01	0.00%	0.01	0.00%	0.01	0.00%
Am-242	0.03	0.00%	0.03	0.00%	0.03	0.00%	0.03	0.00%	0.03	0.00%
Am-243	**21.84**	**0.83%**	**21.34**	**0.85%**	**26.96**	**0.95%**	**32.44**	**1.04%**	**37.80**	**1.13%**
Cm-242	0.74	0.03%	0.68	0.03%	0.69	0.02%	0.69	0.02%	0.69	0.02%
Cm-243	0.17	0.01%	0.11	0.00%	0.11	0.00%	0.11	0.00%	0.11	0.00%
Cm-244	6.07	0.23%	2.91	0.12%	2.91	0.10%	2.91	0.09%	2.91	0.09%
Cm-245	0.56	0.02%	0.55	0.02%	0.70	0.02%	0.84	0.03%	0.98	0.03%
Cm-246	0.12	0.00%	0.12	0.00%	0.15	0.01%	0.18	0.01%	0.21	0.01%
All HM	2629.98	100%	2506.45	100%	2839.56	100%	3111.20	100%	3340.88	100%
All FPs[1]	194.89	7.41%	123.34	4.92%	123.49	4.35%	123.64	3.97%	123.79	3.71%
FPs - Cs,Sr and Daughters[1]	0.76	0.03%	0.65	0.03%	0.80	0.03%	0.95	0.03%	1.10	0.03%

[1]Fractional contribution is relative to the integrated decay heat of all heavy metal nuclides.

III. EVALUATIONS OF TRANSURANIC RECYCLING APPROACHES IN LWRS

In this study, it was assumed that the recycled material originated from spent UO_2 irradiated in a commercial PWR. The plutonium, neptunium, and americium were assumed to be recycled in a similar system a finite number of times, using a MOX, CORAIL (heterogeneous MOX and UO_2), or IMF fuel assembly. The isotopic vector of Pu+Np+Am in the charged assembly in recycle N was derived solely from discharged assemblies in recycle N-1. Blending of recycled and reactor-grade (from spent UO_2) TRU can also be envisioned; however, the current approach is the most straightforward and predicts the maximum benefit achievable for a given number of recycles. The number of recycles before final disposal of the material not consumed in the recycling campaign (N) was considered to be a variable parameter.

In the MOX and IMF cases, the mass of TRU charged in the fresh recycled assembly was adjusted to meet the same operational requirements (full-power days of irradiation) in each recycle. Consequently, the mass of spent fuel processed to produce a given assembly varied from one recycle to the next. In the 17x17 CORAIL assembly, 84 fuel pins on the assembly periphery are MOX, while the interior 180 pins are UO_2. For this case, the Pu+Np+Am from one discharge assembly in recycle N-1 (both UO_2 and MOX pins were reprocessed) was used to fabricate the MOX pins for one new assembly in recycle N, and the U-235 enrichment in the UO_2 pins was adjusted to meet the operational requirements. A more detailed description of the CORAIL fuel cycle can be found in Refs. 3 and 4.

The spent assembly from the N^{th} recycle was assumed to be sent directly to the waste stream. Additionally, the non-recycled minor actinides (curium), the reprocessing waste stream (assumed to be 0.1% of the Np+Pu+Am at each stage), and fission products from each recycle stage (cesium and strontium were removed) were assumed to be disposed in the repository.

This section first provides a description of the neutronics and mass flow analyses which were performed for this study (sub-section III.1). Reactor safety considerations arising from the utilization of transuranic-bearing fuels in a existing LWRs are briefly addressed in sub-section III.2. Lastly, sub-section III.3 provides fuel cycle data for a nuclear enterprise fueled with UO_2 and recycled fuel operating in an equilibrium mode.

III.1. Neutronics and Mass Flow Analyses

Assembly-level calculations were performed assuming a typical 17x17 PWR (UO_2) assembly design, with only substitution of the fuel form in the recycled assemblies (MOX or IMF fuel pins). Assembly design parameters for UO_2, MOX, and IMF assemblies are summarized in Table 3; the design parameters used for the CORAIL assembly were similar to those for the MOX. The heavy metal mass in the MOX or CORAIL fuel assembly is the same as for UO_2, and remains the same for each recycle. Thus, the fuel density and specific power density are constant for all recycles. The concentration of Pu+Np+Am in the fabricated fuel is adjusted with each recycle, however, to provide sufficient reactivity to maintain a discharge burnup of 51 GWd/MTHM. CORAIL cases with discharge burnups of 45 and 51 GWd/MTHM were evaluated.

Table 3. UO_2. Mixed-Oxide, and Inert Matrix Fuel Assembly Design Parameters.

	UO_2, MOX	IMF
Assembly size	17x17 pins	17x17 pins
Number of fuel pins	264	264
Number of guide tubes (GT)	24	24
Number of instrumentation tubes (IT)	1	1
Fuel rod pitch (cm)	1.2598	1.2598
Inter-assembly gap (cm)	0.08	0.08
Fuel pellet material	UO_2 or $(U,TRU)O_2$	Solid solution $(Zr,TRU)O_2$
ZrO_2 hole[1] radius (cm)	N/A	0.2048 (1st recycle) 0.0819 (2nd recycle) 0.0 (3rd recycle)
Fuel pellet radius (cm)	0.4096	0.4096
Clad inner radius (cm)	0.4178	0.4178
Clad outer radius (cm)	0.4750	0.4750
Smeared fuel density (g/cm^3) (pellet at 95% T.D., 1.2% pellet dishing)	9.88	5.60 (1st recycle) 9.00 (2nd recycle) 10.34 (3rd recycle)
Fuel mass (kg HM/assembly)	461.3	42.3 (1st recycle) 347.9 (2nd recycle) 483.1 (3rd recycle)
Zircaloy-4 clad density (g/cm^3)	6.5	6.5
GT/IT inner radius (cm)	0.5715	0.5715
GT/IT outer radius (cm)	0.6121	0.6121
Specific power density (MW/MTHM)	33.69[2]	367.3[2] (1st recycle) 44.7 (2nd recycle) 32.2 (3rd recycle)
Discharge burnup (GWd/MTHM)	51	551 (1st recycle) 67 (2nd recycle) 32[3] (3rd recycle)
Fuel temperature (^oK)	900.0	900.0
Cladding temperature (^oK)	581.0	581.0
Bulk coolant temperature (^oK)	581.0	581.0
Nominal coolant density (g/cm^3)	0.72	0.72

[1]Due to the low thermal conductivity of the ZrO_2 matrix, annular fuel pellets are fabricated with a central region of non-fueled ZrO_2 to limit the centerline temperature.
[2]Based on reactor loading of 193 assemblies and total core power of 3,000 MWth.
[3]In the 3rd recycle, fuel exposure was limited to 1,000 full-power days before reaching reactivity-limited end-of-life. The 1st and 2nd recycles were irradiated for 1,500 full-power days.

For the IMF cases, the heavy metal loading is significantly lower in the first recycle due to the utilization of a non-uranium matrix (factor of 10). With each successive recycle of the unconsumed material, however, the heavy metal (transuranic) loading must be increased to compensate for the reduction of the fissile fraction in the material and to maintain the irradiation cycle length. Consequently, it was necessary to recalculate certain parameters (e.g., fuel density, specific power) needed for the neutronics analysis for each recycle.

As in previous evaluations of transuranic recycling in LWRs, the WIMS8[13] code was utilized for the mass flow analyses. The 172-group, JEF2.2-based cross section library has been previously determined to provide accurate modeling of the important Pu-239,

Pu-240, and Pu-241 resonances. One-eighth-symmetric assembly calculations with reflective boundary conditions were performed, simulating a full-core loading of the UO_2, MOX, CORAIL, or IMF assembly. A linear reactivity letdown model and three-batch irradiation to the discharge burnup were assumed. Consequently, assuming a core neutron leakage of 3.5%Δk, the critical core end-of-cycle condition was approximated by an assembly calculation to 2/3 of the discharge burnup with k_∞=1.035.

The transuranic material initially irradiated in the MOX or IMF fuel was assumed harvested from UO_2 which had been enriched to 4.3 wt.% U-235, irradiated to 51 GWd/MT, and cooled for 5 years. The isotopic vector for the recycled transuranics is provided in Table 4; alternate vectors with curium separation performed at 10 and 20 years cooling are also provided. It is noted that the Am-241 fraction in the transuranic vector after 5 years cooling is slightly above the 3% limit imposed for the French MELOX plant, and this fraction continues to grow as spent fuel reprocessing is delayed. The curium, which was assumed to be disposed in the repository, accounts for ~0.6% of the TRU mass in spent UO_2. As an alternative to permanent disposal, the curium could be fabricated into dedicated targets for LWR transmutation or recycled in a fast spectrum system.

Table 4. Plutonium, Neptunium, and Americium Isotopic Vector in Spent UO_2 (51 GWd/MT).

	Number of Years Cooled		
	5 Years	**10 Years**	**20 Years**
Am241	3.400%	5.682%	8.892%
Am242m	0.007%	0.007%	0.007%
Am243	1.518%	1.518%	1.519%
Np237	4.786%	4.786%	4.790%
Pu238	2.376%	2.284%	2.112%
Pu239	47.924%	47.927%	47.948%
Pu240	22.674%	22.759%	22.890%
Pu241	10.669%	8.390%	5.190%
Pu242	6.646%	6.647%	6.651%

The Pu+Np+Am stream was used to fabricate the recycle fuel, with a two-year post-fabrication interval occurring before charge into the reactor. The length of this interval is somewhat important, as ongoing decay of Pu-241 ($t_{1/2}$=14.4 years) to Am-241 reduces the fissile content before irradiation begins, and requires a higher transuranic loading in the fabricated fuel to meet the irradiation cycle requirements. In the MOX and CORAIL cases, the assembly was irradiated to a discharge burnup of 51 GWd/MTHM, and the same discharge burnup was assumed for each subsequent recycle. At discharge, the MOX fuel was cooled for five years, and then reprocessed to harvest the remaining neptunium, plutonium, and americium. The recovered material (0.1% was assumed lost to the reprocessing waste stream) was recycled, with an appropriate increase in the transuranic loading in the next recycle assembly to maintain the fuel discharge burnup.

For the IMF case, the fuel discharge burnup was allowed to vary with each recycle as the heavy metal loading per assembly was adjusted (see Table 3). Rather than fix the discharge burnup, it was appropriate to fix the irradiation cycle length at 500 days, which

gave a total irradiation period of 1,500 full-power days. This is comparable to that of the MOX fuel (1514 days), and gives roughly the same energy produced per assembly. Likewise, the length of the post-fabrication (two years) and post-irradiation cooling (five years) periods were the same as for the MOX fuel.

III.1.a. MOX Recycling Campaign

In the homogeneous MOX cases, the transuranic material was recycled up to five times (each recycle accumulating an additional 51 GWd/MTHM burnup). While it was known beforehand that the campaign was feasible from a neutronics standpoint, reactor operations considerations (e.g., safety coefficients, fuel handling) might preclude the utilization of such extensively recycled fuel.

Figure 1 illustrates the evolution of the MOX fuel transuranic loading and transuranic vector with each recycle stage. The required transuranic loading increases from 17.4 wt.% to 54.4 wt.% from the first to the fifth recycle. This is due to the gradual reduction of the concentrations of the fissile nuclides (Pu-239 and Pu-241) and increase of the absorbing nuclides (Pu-238, -240, -242, Am-241). The source of the depleted uranium matrix for all MOX recycle stages was assumed to be from spent UO_2, with an isotopic vector of 0.02% U-234, 0.82% U-235, 0.61% U-236, and 98.55% U-238 (% by weight). It has been confirmed that there is enough depleted uranium in the spent UO_2 to provide the matrix material through all five MOX recycles.

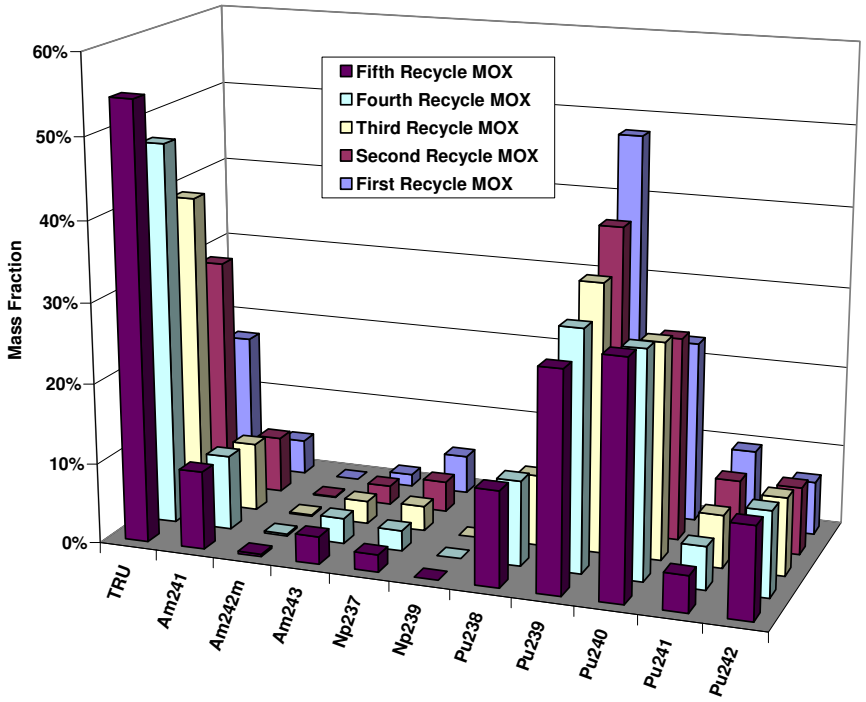

Figure 1. Transuranic Loading (%Pu+Np+Am/HM) and Transuranic Vector in MOX Fuel at Reactor Charge.

Figures 2 and 3 provide the mass of plutonium and other transuranic nuclides during the MOX recycling campaign, normalized to an initial mass of 1 metric ton of UO_2. Thus, these curves show the effectiveness of recycling Pu, Np, and Am in MOX fuel for

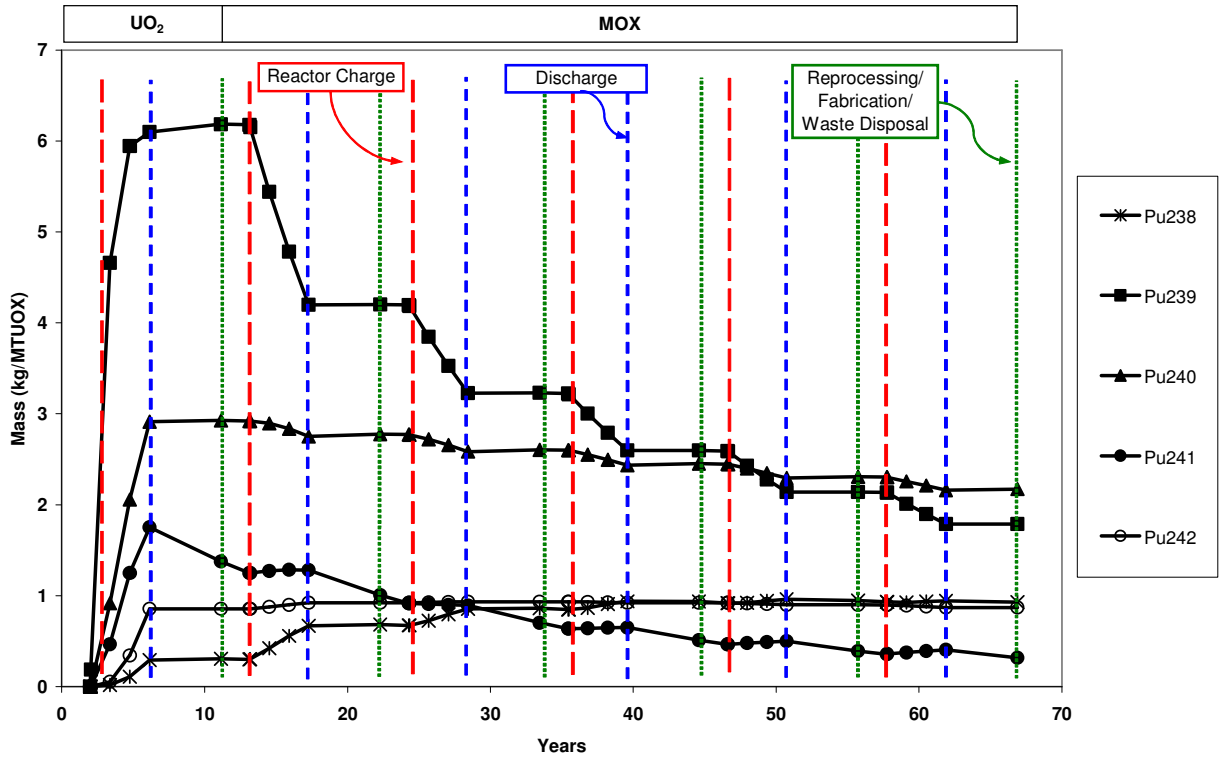

Figure 2. Plutonium Nuclide Masses with Recycling in MOX Fuel. Masses normalized to initial mass of one metric ton UO_2.

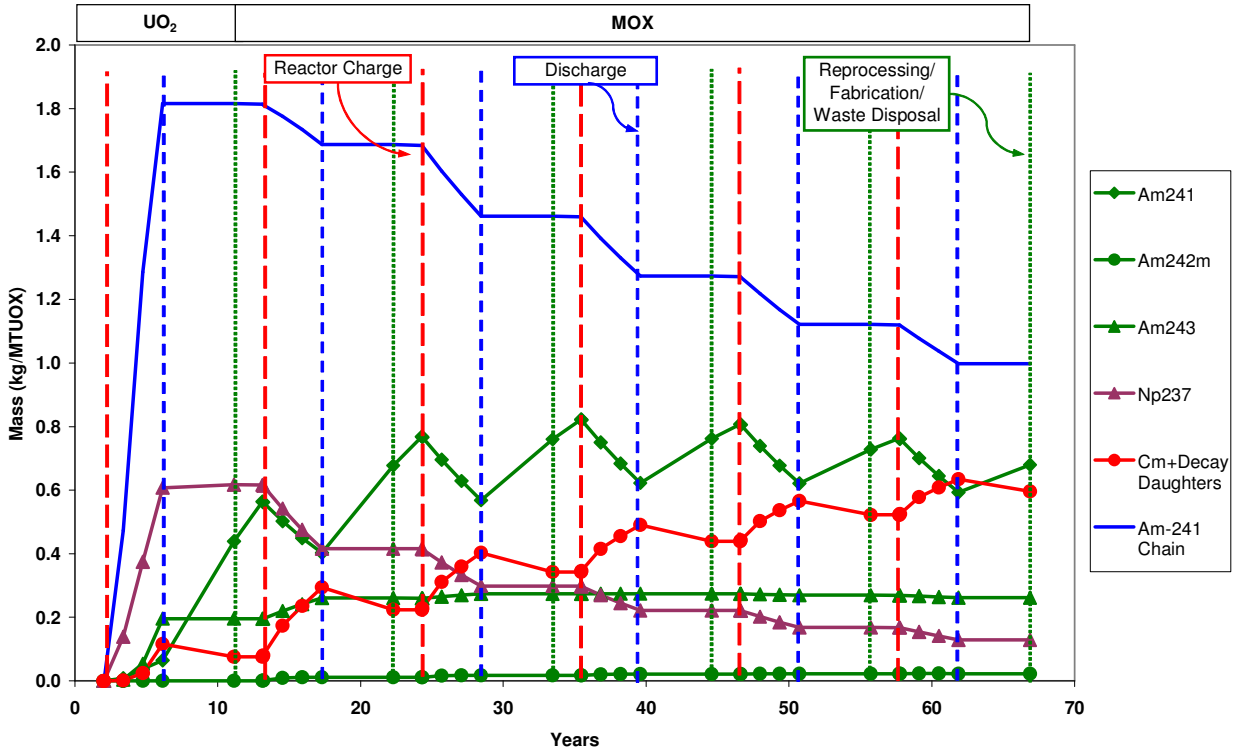

Figure 3. Minor Actinide Nuclide Masses with Recycling in MOX Fuel. Masses normalized to initial mass of one metric ton UO_2.

transuranic consumption, and the mass of each nuclide disposed in the repository at the end of the campaign, relative to direct disposal of spent UO_2 from which the transuranics were initially derived. It should be noted that the small masses of Pu, Np, and Am in the reprocessing waste streams are not included in these curves.

Through five recycles in MOX, 71% of the Pu-239 and Pu-241, and 79% of the Np-237, are consumed. On the other hand, Figure 2 shows that there is a factor of 3 increase in the Pu-238 mass, and according to Figure 3 there is a factor of 6 increase in the mass of curium and its decay daughters disposed in the repository relative to the once-through UO_2.

Figure 4 illustrates the transmutation chains from Np-237 and Am-241, both of which are readily converted to Pu-238. (The one-group absorption cross sections for Np-237 and Am-241 are comparable to those of other actinides in the LWR spectrum.) Since the Pu-238 contributes a significant fraction of the disposed actinide decay heat at the time when forced-air cooling of the facility is turned off (at ~100 years), increasing the mass of Pu-238 in the disposed waste will reduce the repository benefit seen from this recycling campaign. It is important to note that the increase in the Pu-238 concentration is equally attributed to the recycling of Np-237 and Am-241.

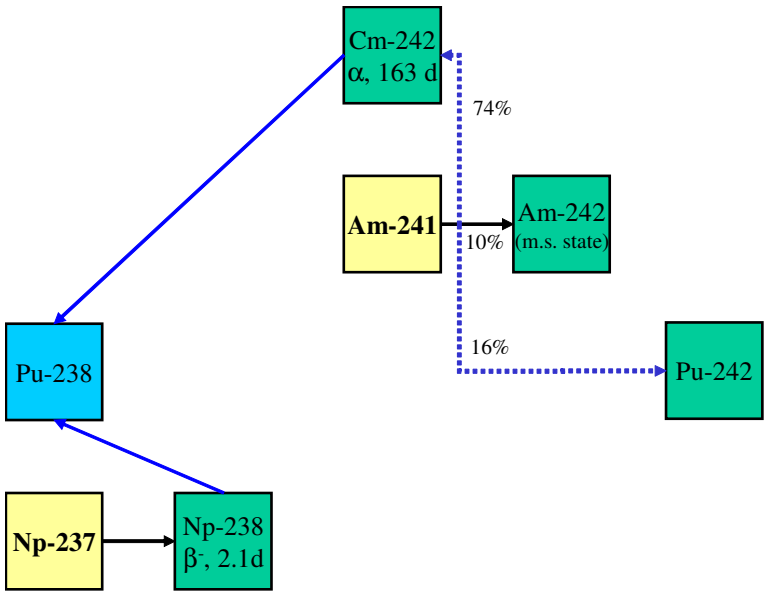

Figure 4. Np-237 and Am-241 Transmutation Chains. Dashed lines represent substitute decay from short-lived Am-242 ground state ($t_{1/2}$=16 hours).

While the curium itself will contribute very little to the overall repository heating (see Table 2), if this waste material is disposed in the repository (the assumption made in this study), the Pu-240 produced from Cm-244 α-decay ($t_{1/2}$=18.1 years) will reduce the potential benefit of the MOX recycling campaign. The magnitude of this penalty has not been precisely calculated, but it is estimated that if the curium is sent directly to permanent disposal the repository loading benefit is reduced by 0.5%-1.0% for each additional MOX recycle.

The Am-241 in the disposed waste form is the most significant contributor to the repository heating in the time frame of interest (100-1500 years after spent fuel

discharge). Both Pu-241 and Am-241 (the nuclides in the so-called Am-241 chain) are readily consumed (transmuted) during fuel irradiation periods through neutron absorption. At the same time, Pu-241 is produced from neutron capture in the recycled Pu-240 at about the same rate at which it is destroyed, so that there is essentially no net consumption of Pu-241 during the irradiation periods (see Section IV.1). The ongoing decay of Pu-241 to Am-241 during the post-irradiation and post-fabrication cooling periods does not alter the Am-241 chain mass, but does yield the "saw-tooth" behavior for the Am-241 mass shown in Figure 3. After the third recycle, the Am-241/Pu-241 ratio in the MOX fuel increases to the point where the Am-241 destruction over a complete recycle (from one discharge to the next) exceeds its production from Pu-241 decay, and through five recycles there is a net consumption of 45% of the Am-241 chain initially harvested from the spent UO_2. This will prove beneficial to the repository in terms of increased loading capacity. It is important to note, however, that the full benefit will not be realized until nearly 70 years of reactor operations with this recycling strategy have been completed.

III.1.b. CORAIL Recycling Campaign

Transuranic recycling in the heterogeneous CORAIL assembly is similar in principle to a partial core loading of homogeneous MOX assemblies in a predominantly UO_2-fueled PWR core. The primary distinction from the MOX case as it was evaluated here (effectively, a full-core loading of MOX) is that the recycled CORAIL assembly contains fresh low-enriched uranium pins, which means that a lower enrichment of transuranics can be used in the MOX pins while meeting the same operational requirements (cycle length). Also, the recycled material is harvested from both the spent UO_2 and MOX pins in the CORAIL assembly from the previous recycle. This slows the degradation of the transuranic vector since reactor-grade plutonium (~60 wt.% fissile) and minor actinides are blended with the Pu+Np+Am derived from the spent MOX pins.

In a study performed in FY02, it was found that transuranic recycling in a PWR core fully-loaded with CORAIL assemblies is capable of reaching an equilibrium state in which the transuranic inventory in the fuel cycle is stabilized and waste disposed to the repository is limited to reprocessing losses and fission products, all the while maintaining reactor safety coefficients similar to those observed for a full-core loading of UO_2 assemblies.[4] The distinctions between that work and the present analysis are that 1) curium is not recycled, which reduces the negative consequences of the buildup of the higher actinides on fuel handling, and 2) only a relatively few number of recycles are considered in this study to evaluate the impact of a short-term transmutation campaign in LWRs.

The CORAIL scenario was first evaluated assuming discharge burnups for the UO_2 and CORAIL assemblies of 45 GWd/MTHM. The homogeneous UO_2 assembly enrichment (0^{th} recycle) for this strategy was 3.82 wt.% U-235, compared with 4.3 wt.% if the discharge burnup is extended to 51 GWd/MTHM. Figure 5 illustrates the impact of subsequent Pu+Np+Am recycling on the CORAIL assembly UO_2 and MOX pin enrichments, and the fissile content of the recycled TRU vector. Also included is the fissile content of the recycled TRU for the homogeneous MOX assembly (see the previous section).

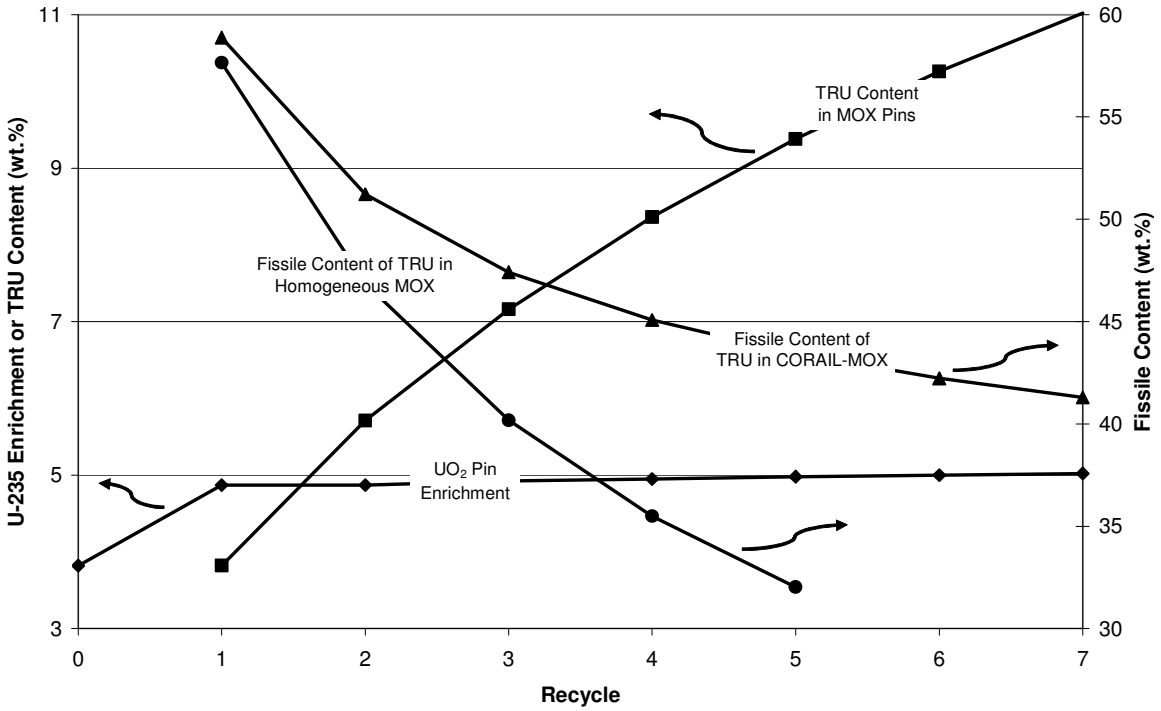

Figure 5. Charged UO_2 and CORAIL Assembly Properties in CORAIL Recycling Strategy. Discharge burnup = 45 GWd/MTHM.

The CORAIL recycling strategy was formulated so that all the Pu+Np+Am harvested from a single assembly in the previous recycle (less reprocessing losses) was used to fabricate the CORAIL-MOX pins in the next recycle stage. Following this strategy yielded a MOX pin loading of 3.82%TRU/HM in the first recycle (it is only coincidental that this is the same as the homogeneous UO_2 enrichment). In order to meet the reactivity-limited end-of-life burnup of 45 GWd/MT, it was necessary to enrich the UO_2 pins in the interior of the CORAIL assembly to 4.87 wt.% U-235. With each successive recycle, the TRU content in the MOX pins increases as more Pu+Np+Am is produced in the UO_2 pins than is consumed in the MOX pins (although the rate of increase is slowing as the equilibrium state is being approached). By the 7th recycle, the MOX pin TRU content has reached 11.0% and the UO_2 pin enrichment must be increased to 5.0% to meet the cycle length requirements. The soluble boron worth and moderator temperature coefficient at this stage were evaluated and found to be comparable to those for the conventional UO_2 assembly. It is noted that since the recycled TRU is derived from both the MOX and UO_2 pins in the CORAIL assembly, the TRU fissile content declines much slower than for recycling in homogeneous MOX assemblies, which was not blended with reactor-grade Pu+Np+Am (see Figure 5).

Through five recycles of the Pu+Np+Am in CORAIL assemblies discharged at 45 GWd/MT, there is a net consumption of 70% of the Pu-239 and ~60% of the Pu-240 and Pu-241 which would have been sent to the repository if the spent UO_2 assembly was directly disposed. The net consumption of the Am-241 chain is 53%, compared with 45% consumed through the same number of recycles of the homogeneous MOX (discharged at 51 GWd/MT); increasing the discharge burnup of the CORAIL assemblies to 51 GWd/MT will further increase the Am-241 chain consumption. Likewise, the Pu-

238 mass sent to the repository through 5 CORAIL recycles is doubled relative to the once-through fuel cycle, whereas there is a factor of 3 increase in the Pu-238 mass in the homogeneous MOX strategy. Thus, it is anticipated that the CORAIL recycling strategy will be more beneficial to the repository than the homogeneous MOX strategy for a given number of recycles.

The CORAIL mass flow analyses were repeated assuming a discharge burnup 51 GWd/MTHM to facilitate future inter-comparisons of the MOX, CORAIL, and IMF results. So far, only calculations for the first CORAIL recycle have been performed to assess the impact of the higher discharge burnup. As the UO_2 discharge burnup increases from 45 to 51 GWd/MT, the concentration of transuranics in the spent fuel increases. The larger mass of Pu+Np+Am harvested from the spent UO_2 assembly increases the TRU content of the MOX pins in the first recycle CORAIL assembly from 3.82 wt.% to 4.09 wt.%. Although the higher TRU loading in the MOX pins adds some reactivity to the CORAIL assembly, it is not enough to compensate for the increased discharge burnup of the CORAIL assembly. Consequently, the enrichment of the UO_2 pins in the first CORAIL recycle must be increased from 4.87 wt.%U-235 to 5.53%, and will climb higher as recycling continues with this strategy. A re-evaluation of this strategy may be necessary as this enrichment exceeds the conventional limit on fabrication of low-enriched uranium fuel.

III.1.c. IMF Recycling Campaign

In addition to the analysis with the MOX fuel form ((depleted-U,Pu,Np,Am)O_2), the impact of recycling the Pu+Np+Am in an inert-matrix fuel (IMF) form was also considered. Again, the source of the transuranic material was spent UO_2 irradiated to 51 GWd/MT. The inert-matrix fuel form considered here was solid-solution (Zr,Pu,Np,Am)O_2. Due to the lower thermal conductivity of the zirconia relative to typical MOX or UO_2, an annular fuel pellet design was assumed so that the fuel centerline temperature would be within a reasonable limit.[14] A separate study that systematically compares solid-solution and dispersion-type inert-matrix fuel forms for their transmutation and operations performance will also be completed this year.

Without fertile U-238 in the inert-matrix fuel, no additional transuranics are produced during irradiation and a significant net consumption of the transuranic material is achieved. Inert-matrix fuels were initially envisioned as part of the weapons-material disposition program, as they target the consumption of Pu-239. This is illustrated in Figure 6, which shows the isotopic vector of the material loaded in the IMF fuel with each recycle. However, this characteristic of IMF will actually limit its potential benefit to the repository since the number of recycles will be constrained by the declining reactivity of the recycled material. If a constant irradiation cycle length is to be maintained, it is likely that only a few recycles will be possible.

Figure 7 compares the k_∞ with burnup for UO_2 and IMF assemblies. The IMF assembly has an initially shallower reactivity letdown, similar to that typically seen for MOX fuel, due to the harder spectrum which is typical for TRU-bearing fuels. In order to maintain the 500-day cycle length in the second recycle stage, it was necessary to increase the total transuranic loading per assembly by a factor of 8 to compensate for the significant plutonium burnout; this was accomplished by reducing the size of the center

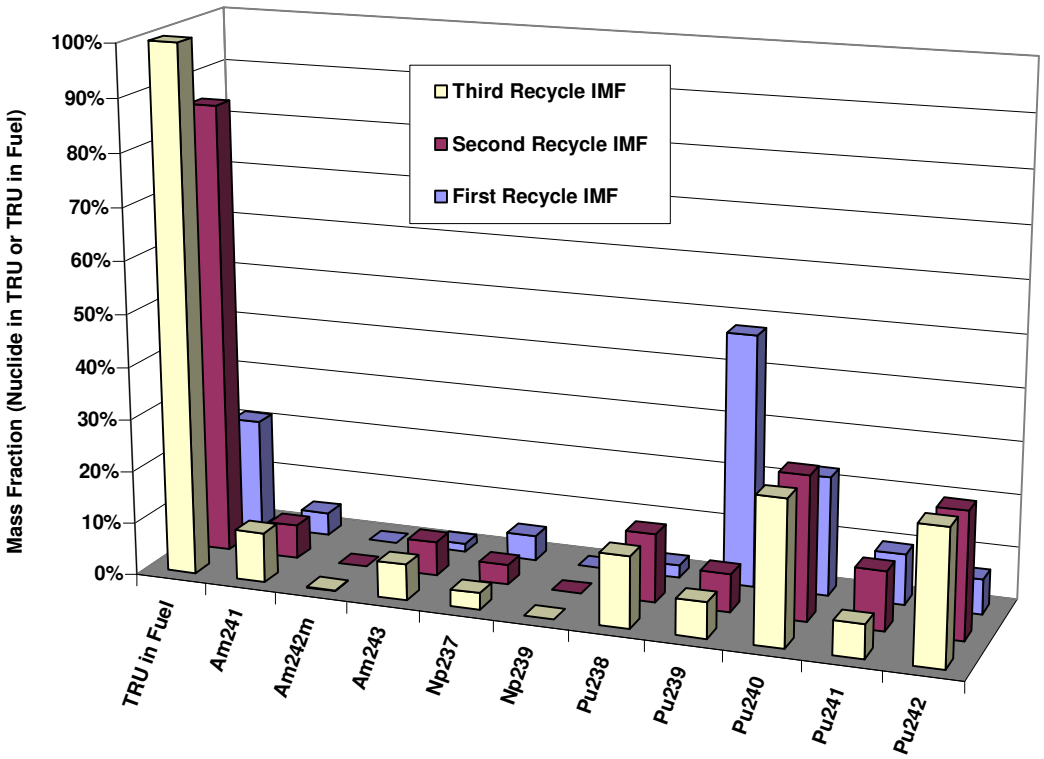

Figure 6. Transuranic Loading (%Pu+Np+Am) and Transuranic Vector in IMF Fuel at Reactor Charge.

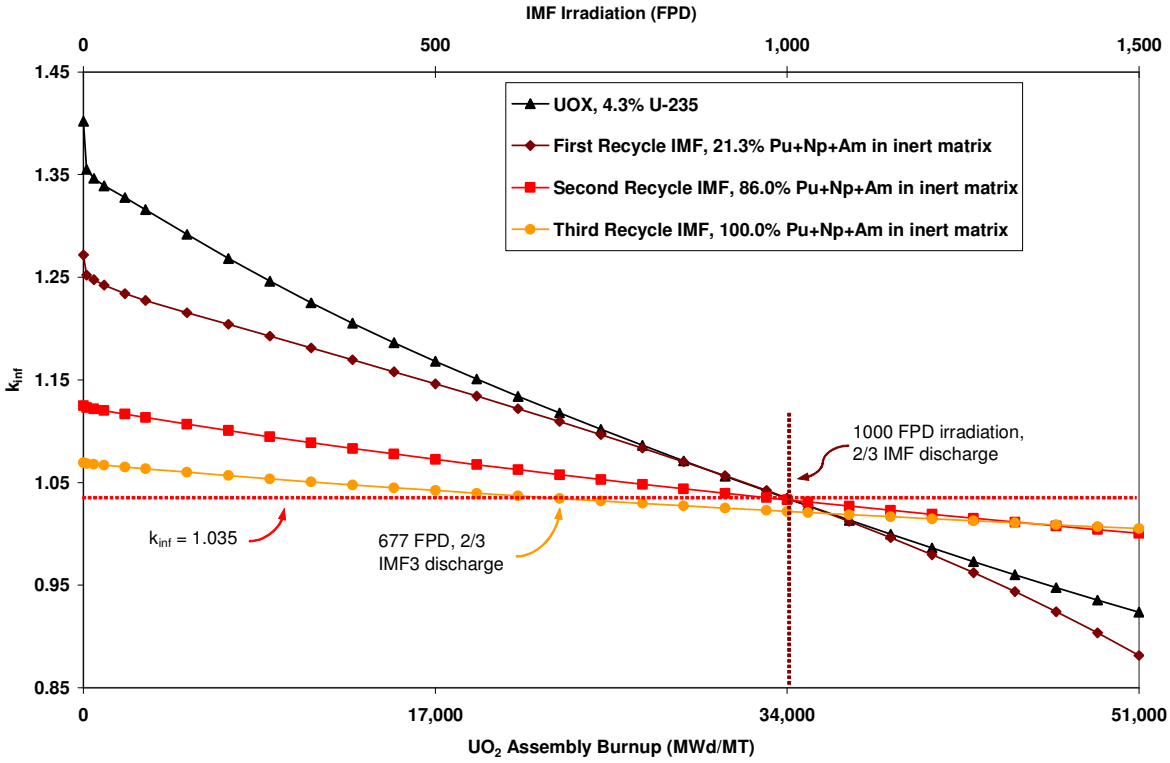

Figure 7. Assembly Eigenvalue for Inert-Matrix Fuel Recycle Cases.

zirconia hole in the fuel pin, as well as increasing the TRU/zirconia mass ratio in the fuel. By the third recycle, fabricating the fuel pins with 100% (Pu,Np,Am)O_2 (the zirconia hole and all of the zirconia matrix were eliminated), and assuming a core fully-loaded with such assemblies, yielded a maximum cycle length of 333 days (1000 full-power days irradiation). Consequently, the recycling campaign in the IMF case must be terminated after only three recycles; practically, no more than two recycles would be considered.

The transuranic mass in the first recycle IMF case is 42.3 kg/assembly, compared with 80.2 kg/assembly for first recycle MOX. The reason for this reduction is the elimination of the depleted-uranium matrix and neutron capture in U-238. In the first recycle MOX assembly, the U-238 neutron capture rate is comparable to the capture rates in Pu-239 and Pu-240. Furthermore, as shown in Figure 8, reducing the assembly transuranic loading softens the neutron spectrum relative to the MOX case, which increases the fission cross sections and lowers the fissile transuranic loading needed to meet the specified power normalization.

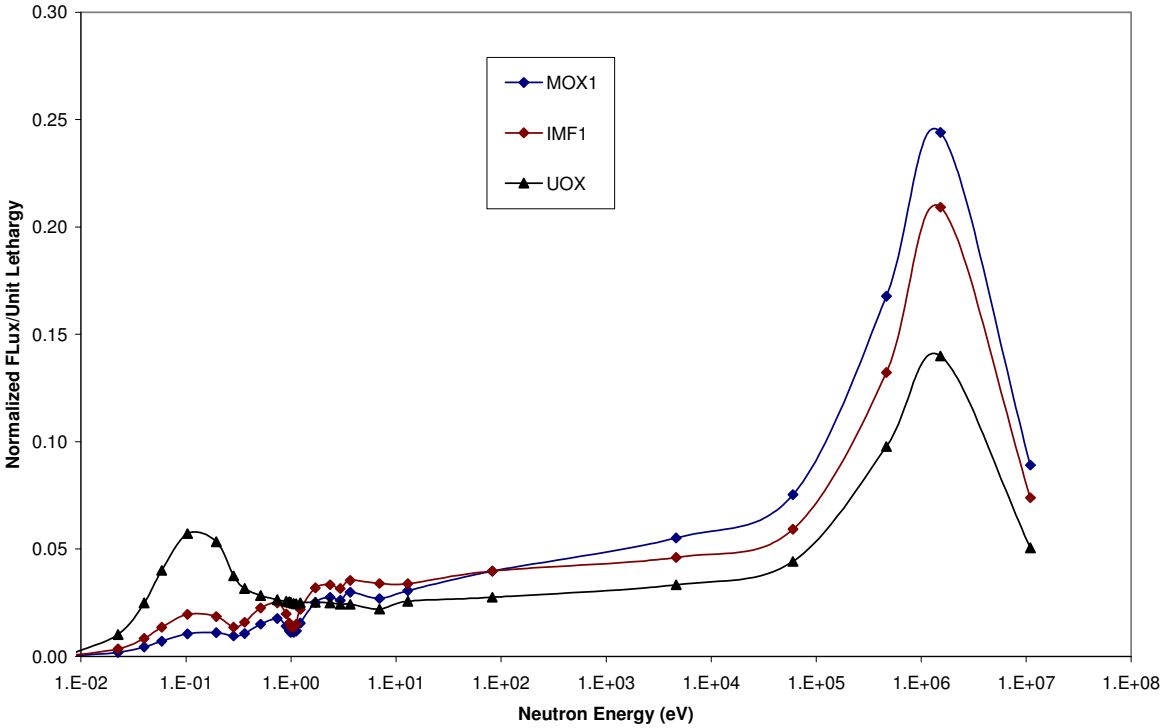

Figure 8. Assembly Averaged Neutron Spectra in UO2, and First Recycle IMF and MOX Assemblies. Beginning-of-life, 0 ppm soluble boron.

Figures 9 and 10 provide the plutonium and minor actinide nuclide masses relative to 1 metric ton of UO_2 for a Pu+Np+Am recycling campaign utilizing inert-matrix fuel. The net consumptions of Pu-239 and Pu-241 through three recycles are 95% and 80%, respectively. Also, there is a net consumption of 62% of the Am-241 chain nuclides which would be present in direct-disposed UO_2. Furthermore, with fewer number of recycles, the mass of Pu-238 disposed to the repository relative to direct-disposed UO_2 is increased by only a factor of 2, compared with a factor of 3 increase through five recycles achieved in the MOX fuel case. Thus, a greater repository benefit with fewer recycles is

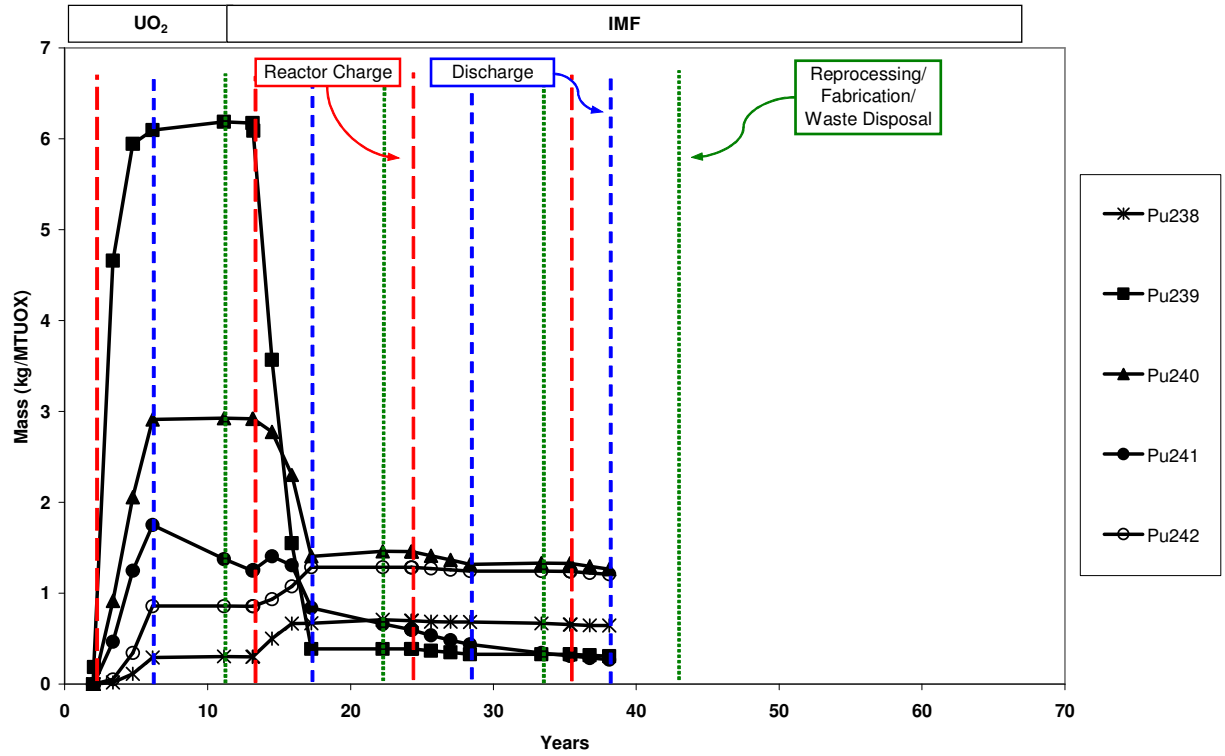

Figure 9. Plutonium Nuclide Masses with Recycling in Inert-Matrix Fuel. Masses normalized to initial mass of one metric ton UO_2.

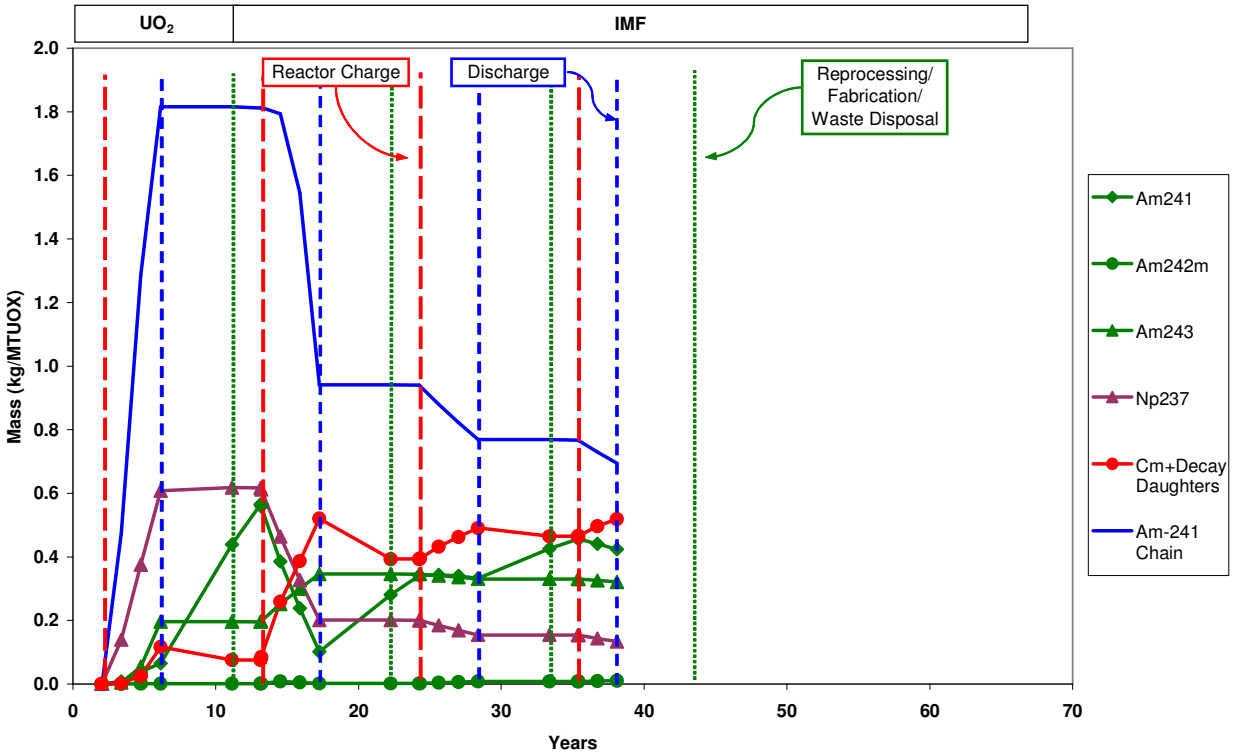

Figure 10. Minor Actinide Nuclide Masses with Recycling in Inert-Matrix Fuel. Masses normalized to initial mass of one metric ton UO_2.

expected for the IMF case, although this campaign depends on the development of a fuel form which has not yet been utilized for commercial power production.

III.2. Reactor Safety Considerations

The utilization of a full-core loading of transuranic-bearing fuels will alter the reactor response to transients compared with conventional UO_2. Particular concerns about the coolant void effect arise from the high transuranic loadings considered here. The hardened spectrum accompanying coolant voiding increases the neutron production/absorption (η) in Pu-239, Pu-240, and Pu-241, as well as reduces the parasitic capture in Np-237 and Am-241, potentially resulting in a positive reactivity insertion.

Figure 11 illustrates the effect of coolant voiding on the assembly k_∞ for a number of cases. For UO_2, the void effect is negative over the entire range of voiding due to the utilization of an undermoderated assembly design. For first recycle MOX and IMF assemblies with the same design parameters, the void effect is negative up to about 60% coolant voiding, but becomes positive as the voiding fraction continues to increase.

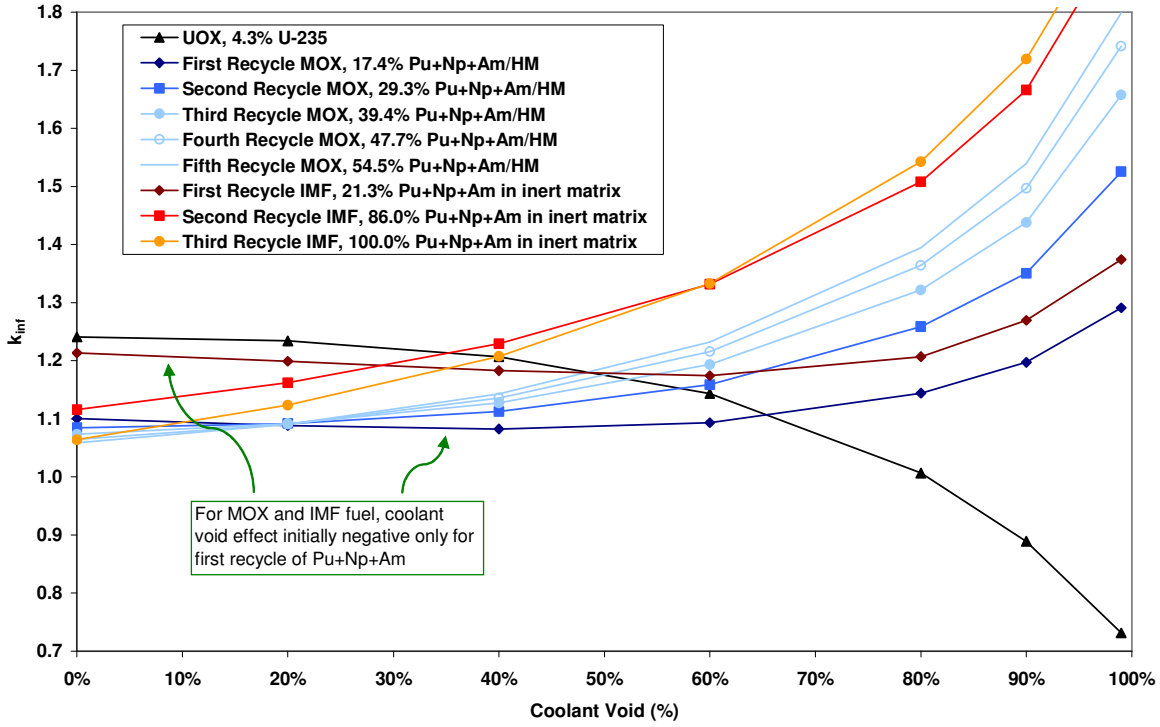

Figure 11. Effect of Coolant Voiding on Assembly k_∞. Beginning-of-life, 1500 ppm soluble boron.

For all other recycles, the reactivity response is positive for even the smallest amount of coolant voiding, although the response is somewhat exaggerated because the lattice model with reflective boundary conditions used here neglects the increased neutron leakage (a negative reactivity effect) which occurs with coolant voiding. Coolant void worth calculations (again, neglecting leakage effects) were previously performed for the CORAIL case and found to be comparable to that of a conventional UO_2 assembly, even up to a near-equilibrium TRU loading of 20%.[4]

It is, however, unlikely that full-core loadings of the types of assemblies considered here will be utilized in practice. Rather, a core loaded with a heterogeneous mixture of UO_2 and MOX or IMF assemblies could be used to reduce the impact of the coolant voiding effect. For the recycling campaigns with MOX and IMF considered here, the fraction of UO_2 assemblies in the nuclear enterprise (from which the transuranic material originates) is ~85% (see Section III.3 below).

While the mass flow results presented here are based on the simulation of a full-core loading (assembly-level calculations with reflective boundary conditions), it can be seen from Figure 12 that the depletion characteristics of an IMF fuel pin are relatively insensitive to the composition of neighboring pins. The results for the IMF "pin" in Figure 12 are from a WIMS8 lattice calculation of 1 IMF pin surrounded by 8 UO_2 pins (89% UO_2 in the lattice), while the "hom(ogeneous)" depletion results are based on an assembly-level calculation with all IMF pins. Similar results have been observed for the case of a MOX assembly, depleted in either a heterogeneous lattice (1/4-core loading of MOX in a predominantly UO_2- fueled core) or based on an assembly-level calculation. Thus, the LWR or ALWR cores could be loaded with a small fraction of MOX or IMF assemblies (or even pins in a heterogeneous assembly layout such as CORAIL) and achieve roughly the same results as predicted here with regard to net transuranic consumption. At the same time, this would alleviate the negative impact of TRU-bearing fuel on the whole core reactivity coefficients.

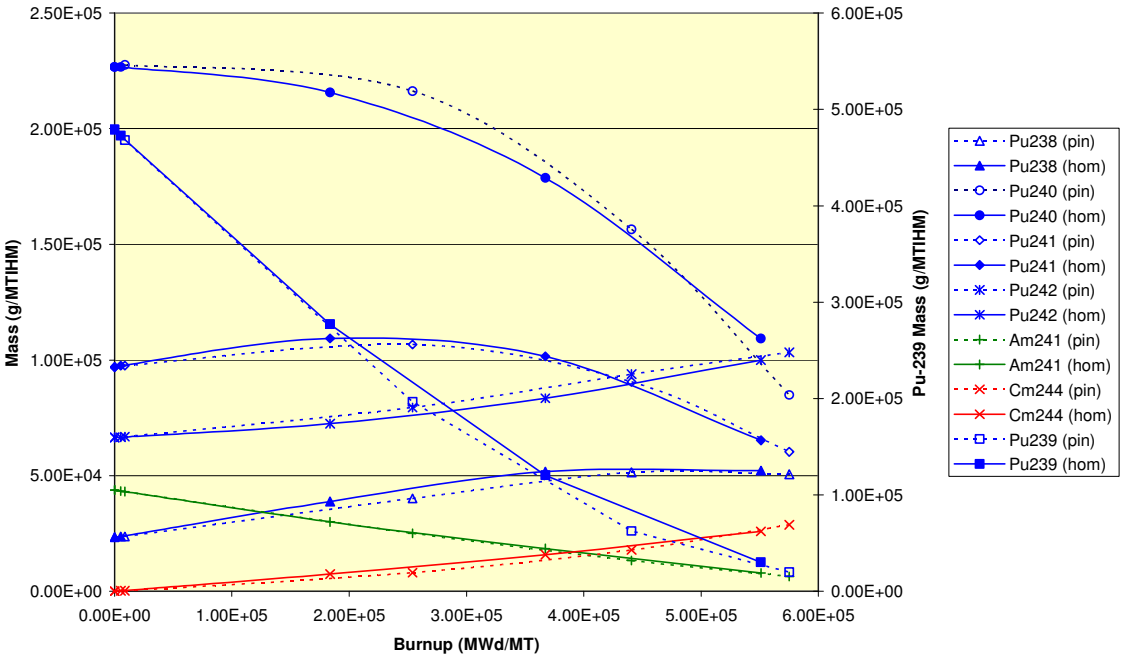

Figure 12. Comparison of Nuclide Masses in IMF Pin Surrounded by UO_2 (pin) and in Homogeneous IMF Assembly. First recycle of Np+Pu+Am derived from commercial spent nuclear fuel.

III.3. Nuclear Enterprise Data

In addition to the assembly-level calculations which were performed to determine the spent fuel assembly isotopics, an evaluation of the commercial nuclear enterprise and associated infrastructure requirements was performed for the three LWR recycling strategies considered here. The nuclear enterprise is envisioned as consisting of a mix of reactors fueled predominantly with UO_2, and MOX, CORAIL, or IMF fuel assemblies with recycled material. Each reactor in the enterprise could be either homogeneously loaded with a single assembly type (in which case there would be some units with a full-core loading of TRU-bearing fuel) or, as discussed above, a given reactor could be heterogeneously loaded with UO_2 and recycled fuel. The latter approach would alleviate some of the difficulties associated with burning TRU-bearing fuel in reactors with safety systems designed for low-enriched uranium fuel.

For each of the recycle strategies, the material disposed in the repository consists of reprocessing wastes and directly disposed spent fuel assemblies. The relative amounts of UO_2 and recycled fuel loaded in the nuclear enterprise are needed in order to properly calculate the repository heating arising from the disposed waste. For this study, an "equilibrium" enterprise was assumed, that is, just enough UO_2 is burned in the system to produce the Pu+Np+Am needed to fuel all "downstream" assemblies through the N^{th} recycle. Effectively, all newly-fabricated UO_2 in the enterprise is reprocessed, but none of the spent fuel in the "legacy waste" is utilized.

The "F_l" values provided in Table 5 are the amount of fuel (*initial* heavy metal) from recycle l that must be reprocessed to harvest enough transuranic material to fabricate a given amount of fuel material for recycle l+1. So, for example, the spent fuel associated with 13.5 MTIHM UO_2 must be reprocessed to harvest enough Pu+Np+Am to fabricate 1 MTIHM of the first-recycle MOX (174 kg TRU/MTIHM in this case). Subsequently, the spent fuel from 1.98 MTHIM of the first-recycle MOX must be reprocessed to produce 1 MTIHM of the second-recycle MOX. For the CORAIL case, the problem was formulated so that there is a one-to-one relationship between the spent and recycled assemblies. It should be noted that the balance of the material needed to fabricate 1 MTIHM (either depleted uranium for MOX and CORAIL-MOX pins, or low-enriched uranium for CORAIL-UO_2 pins) must also be available. A significantly larger amount of spent UO_2 must be reprocessed to produce 1 MTIHM of the first-recycle IMF fuel, but it must be remembered that the recycled fuel in this case consists only of transuranics.

Figure 13 illustrates the fraction (by total core volume) of the various fuel types loaded in the nuclear enterprise for each of the recycling strategies considered here. In the CORAIL strategy, there are roughly twice as many MOX pins in the nuclear enterprise, but these have a much lower TRU content (see Section III.1.b). Assuming equal power sharing between the UO_2 and the recycled fuel, the values given here provide a first-guess at the fraction of nuclear power generated by conventional UO_2 and advanced fuel forms (MOX or IMF). Although the assumption of equal power sharing is not necessarily a good one since the recycling strategies have not been optimized, it is clear that as the number of recycles increases, less low-enriched uranium (the balance of the values charted in Figure 13) is needed in the enterprise and a smaller fraction of the total nuclear power is generated by UO_2.

Table 5. Calculated "F_l" Values for MOX, CORAIL, and IMF Recycling Campaigns. F_l is the mass of fuel from recycle l (MTIHM) that must be reprocessed to produce one MTIHM of fuel material for recycle l+1.

l (N=l+1)	MOX	CORAIL	IMF
0 (N=1)	13.50	1	77.57
1 (N=2)	1.98	1	2.42
2 (N=3)	1.52	1	1.10
3 (N=4)	1.35	1	N/A
4 (N=5)	1.26	1	N/A

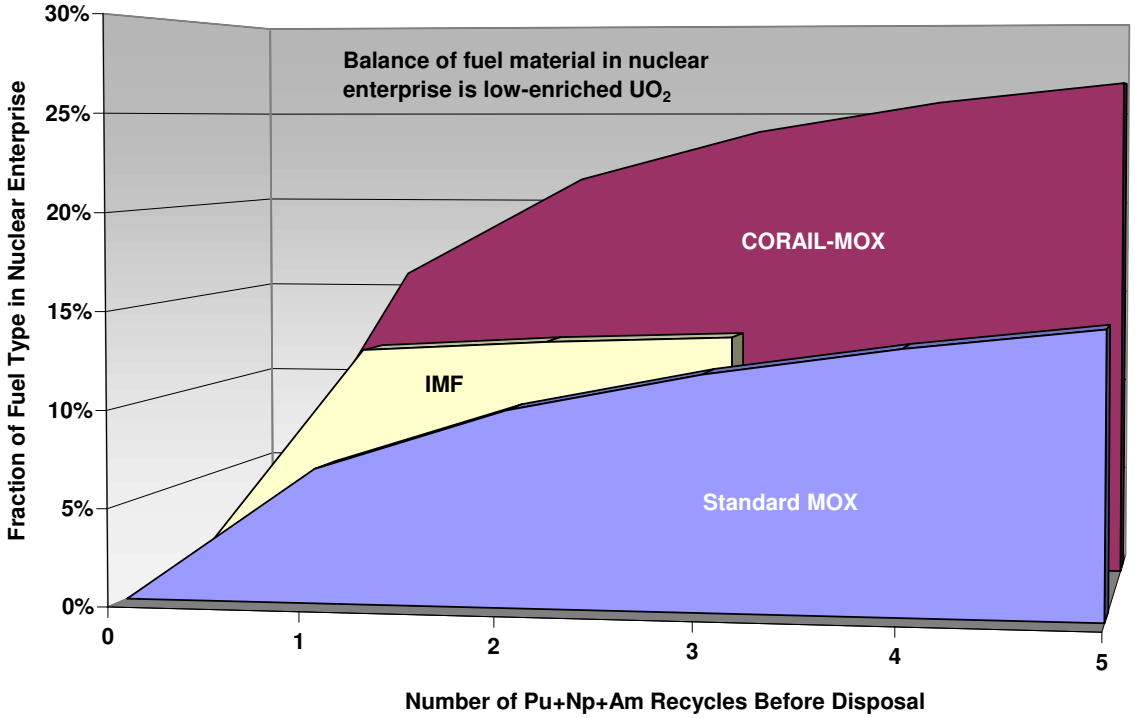

Figure 13. Distribution of Fuel Types in Equilibrium Nuclear Enterprise with MOX, CORAIL, and IMF Recycling Strategies.

Table 6 provides the fuel fabrication and spent fuel reprocessing infrastructure requirements. These values are based on a total capacity of 300 GWth for the nuclear enterprise and a capacity factor of 85%. Values were calculated for the cases of one recycle of Pu+Np+Am before disposal, and five (MOX, CORAIL) or three (IMF) recycles before disposal. For comparison, the conventional once-through fuel cycle requires a fabrication capacity of 1,825 MTIHM/year and no spent fuel reprocessing.

For the MOX and IMF strategies, the mass flow analyses assumed that all fabricated UO_2 is enriched to 4.3 wt.% U-235. In the CORAIL strategy, only the homogeneous UO_2 assemblies are at this enrichment; the UO_2 pins in the CORAIL assembly (denoted "UO_2 (C)" in Table 6) are higher enriched, and the enrichment increases with the recycle stage. Thus, while the CORAIL strategy may require a smaller total UO_2 fabrication

Table 6. Fabrication and Spent Fuel Reprocessing Infrastructure Requirements of 300 GWth Nuclear Enterprise for MOX, CORAIL, and IMF Recycling Campaigns.

	Fabrication Requirements (MTIHM/year)		Spent Fuel Reprocessing (MTHM/year)	
MOX Recycling Strategy				
	N=1	*N=5*	*N=1*	*N=5*
UO_2	1699.1	1561.4	1610.0	1479.5
MOX	125.9	263.6	0.0	228.5
Total			1610.0	1708.0
CORAIL Recycling Strategy[a]				
	N=1	*N=5*	*N=1*	*N=5*
UO_2	912.5	304.0	864.7	1074.4
UO_2 (C)	622.2	1037.1		
MOX	290.3	483.9	0.0	366.9
Total			864.7	1441.3
IMF Recycling Strategy				
	N=1	*N=3*	*N=1*	*N=3*
UO_2	1600.0	1583.8	1516.2	1500.8
IMF	20.8	40.7	0.0	17.1
Total			1516.2	1517.9

[a]CORAIL recycling infrastructure requirements calculated based on 51 GWd/MT discharge burnup.

capacity for a given number of recycles, the annual uranium ore requirements for this strategy will likely be somewhat larger than that needed for the MOX or IMF strategies. As noted in Figure 13, a larger MOX fabrication capacity is needed for the CORAIL strategy, but the TRU content of the MOX is lower by a factor of ~4 (4.1% vs. 17.4% for first-recycle CORAIL-MOX and homogeneous MOX, respectively).

Significant technology development and practical experience with reprocessing spent UO_2 has been achieved worldwide. Reprocessing of spent MOX is technically similar, and is anticipated for the French nuclear program. Reprocessing of inter-matrix fuel forms still requires technology development, but given the limited gain of subsequent recycles of IMF fuel, this may not be an option worth pursuing. The spent fuel reprocessing capacity is the smallest for the single-recycle CORAIL approach, since only half of the enterprise is fueled with conventional UO_2 assemblies. However, in all cases, the reprocessing capacity is no more than 1,700 MTHM/year.

IV. REPOSITORY LOADING BENEFIT FROM TRANSURANIC MANAGEMENT IN LWRS

The benefits of the various recycling strategies considered here, expressed in terms of increased repository capacity, were obtained from thermal analyses of the repository. A detailed description of the repository thermal analyses and the consequent loading benefit resulting from multiple recycles of plutonium, neptunium, and americium in an LWR are provided in Ref. 15. This section provides a brief summary of the thermal analysis. Also, the various recycle fuel forms are compared with regard to their repository loading benefit, and the impact of delaying spent fuel reprocessing and recycling is also discussed. Lastly, the impact of the particular transuranic separations strategy on is evaluated.

The decay heat characteristics of the disposed material, whether reprocessing wastes, fission products, or transuranics not consumed in the recycling campaign, were evaluated with the ORIGEN2 code. One group cross section data for a MOX assembly loaded in a mixed UO_2/MOX core are included with the code package as a standard-use library (the so-called *pwrpupu* library). In order to improve the accuracy of the ORIGEN2 analysis relative to the multi-group WIMS8 results, appropriate substitute cross section data for some actinide and fission product nuclides were derived from the WIMS8 output. For all other nuclides, cross section data from ORIGEN2 *pwrpupu* library were utilized without modification.

For the reference case of spent UO_2 assemblies discharged at 51 GWd/MT burnup and directly disposed in the repository after 25 years post-irradiation cooling, a loading limit of 1.15 MTIHM/m in the drift tunnels has been determined from time-dependent thermal analysis.[15] Alternately, the loading can be expressed in terms of the net thermal energy produced by the spent fuel which is disposed in the repository per meter of tunnel length (GWd/m); this unit is useful for comparing the repository loadings of cases with different initial masses of heavy metal per assembly, such as IMF vs. MOX. For the reference case, the loading limit in the repository expressed in this way is

$$1.15\frac{\text{MTIHM}}{\text{meter}}\cdot 51\frac{\text{GWd}}{\text{MTIHM}} = 58.6\frac{\text{GWd}}{\text{meter}}.$$

This spent fuel loading yields a peak rock pillar temperature of 96°C, which occurs about 1,500 years after the spent fuel is discharged from the reactor. Since the transport of the heat generated in the waste package to the rock pillar will take some time to occur, it is felt that the results in Table 2 for the integrated decay heat from 100 to 1,250 years perhaps provide the best estimate of the contributors to the rock pillar peak temperature for the reference case.

The repository loading as a function of the number of recycles was evaluated based on the decay heat generated by the direct disposal of the assembly in the final ("N^{th}") recycle, as well as disposal of all waste materials accumulated from spent fuel reprocessing in any previous recycles. The reprocessed waste material included all curium, 0.1% of the Pu, Np, and Am, and all fission products except cesium, strontium, and their daughters; the Cs and Sr were assumed sent to temporary storage. In the recycling strategies, spent fuel assemblies are diverted to reprocessing and recycling rather than direct disposal in the repository. Thus, the decay heat generated by the final directly-disposed assembly (and associated reprocessing wastes) substitutes for the heat

which would have been contributed by the reprocessed fuel had it been disposed directly in the repository. In order to account for this, the decay heat of the material finally disposed in the repository was normalized to the "net burnup" of the material in the N^{th} recycled assembly,

$$B_N^{Net} = \sum_{k=0}^{N} \left[B_k \cdot \prod_{l=k}^{N-1} F_l \right], \tag{2}$$

where B_k is the actual burnup of the fuel in recycle k (expressed in GWd/MTHM), F_l is the initial mass of fuel (MTIHM) in recycle l that must be reprocessed to produce 1 MTIHM of fuel in recycle l+1, and N is the total number of recycles practiced. Calculated values for F_l for the MOX, CORAIL, and IMF recycling campaigns are provided in Table 5, above.

IV.1. Impact of Recycle Fuel Form

Figure 14 compares the time-dependent heavy metal decay heat generated by the directly-disposed assembly for a number of cases, normalized to the net burnup of the spent fuel before disposal. The normalized heavy metal decay heat is initially higher in the recycle cases due to the higher discharge masses (per assembly and per GWd) of Cm-244 ($t_{1/2}$=18.1 years) and Pu-238 ($t_{1/2}$=87.7 years) in the directly disposed recycled assembly; although plutonium is recycled to consume it, there is a significant net *production* of Pu-238 in the first one or two recycles from parasitic capture in Np-237 and Am-241 (see Figure 4). After about 200 years cooling, however, the heavy metal decay heat is lower for the recycled fuel due to the beneficial consumption of Pu-239, Pu-240, Pu-241, and Am-241.

The "Repository Loading Benefit" values reported in Figure 14 are taken from detailed thermal analyses reported in Ref. 15, and, with one exception as noted, are based on an assumption of five years post-irradiation cooling before spent fuel processing and curium separation. These values quantify the maximum achievable repository loading capacity for the indicated strategy relative to direct disposal of once-through UO_2. For *a single recycle* of Pu+Np+Am in homogeneous MOX (51 GWd/MT burnup), CORAIL (51 GWd/MT burnup), and IMF (551 GWd/MT burnup) fuel, the repository loading limit is increased by 9%, 22%, and 79%, respectively, relative to once-through UO_2. The repository loading increase is primarily the result of reducing the mass of Am-241 chain nuclides sent to the waste by 13%, 21%, and 54% for the MOX, CORAIL, and IMF cases, respectively, relative to once-through UO_2.

Table 7 provides one-group actinide cross sections at beginning-of-life for UO_2, MOX, and IMF assemblies. There is a significant production path for Pu-241 during fuel irradiation from neutron capture(s) in Pu-240, Pu-239, and, in the case of MOX fuel, U-238. In fact, the gross production of Pu-241 during MOX irradiation is equivalent to that initially loaded in the assembly, while in an IMF assembly the gross production is 3 times the initial loading. It is also noted, however, that the absorption cross section for Pu-241 in the recycled (TRU-bearing) fuel forms is the largest of all the plutonium isotopes, and the Am-241 absorption cross section in fresh MOX and IMF assemblies is only about 5% lower than that of Pu-241. Consequently, the destruction of the Am-241 chain nuclides through TRU recycling in an LWR is relatively efficient.

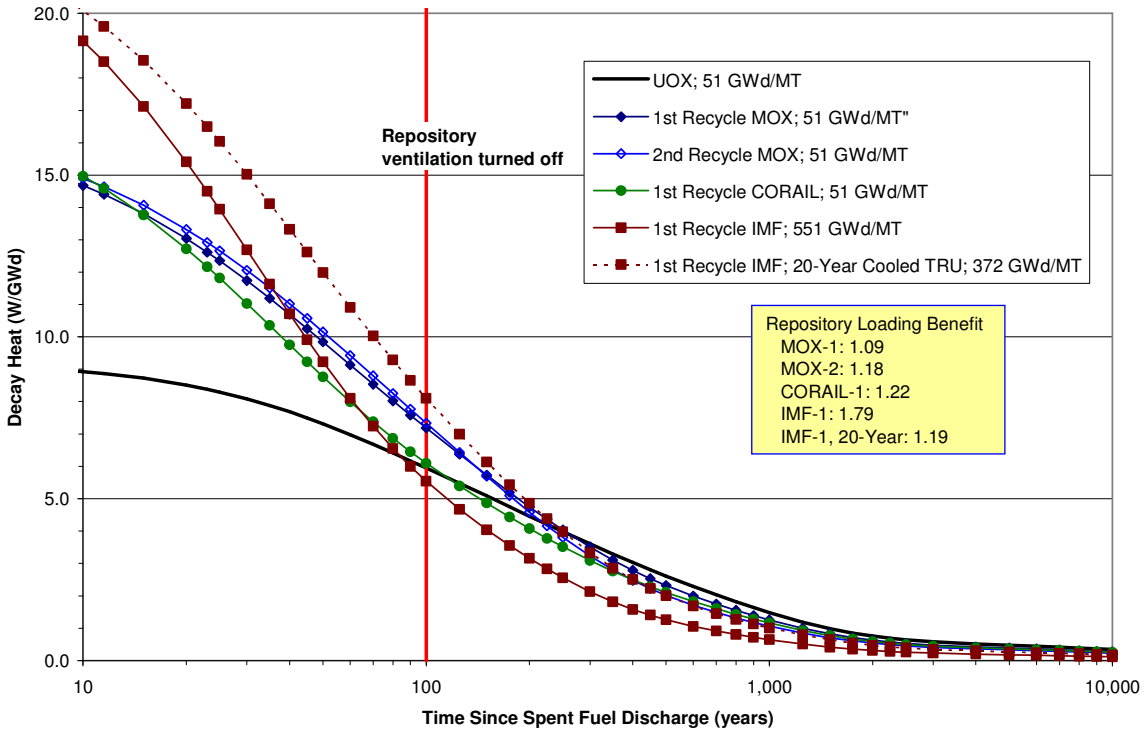

Figure 14. Heavy Metal Decay Heat in Direct-Disposed Spent Fuel Assembly/Net Energy Produced. Recycled transuranics with curium separation at 5 years, unless indicated otherwise.

Figure 15 compares the Pu-241 production (Pu-240 neutron capture rate) and destruction rates (from absorption and decay) in MOX and IMF assemblies fabricated with 5-year cooled TRU. To facilitate comparison, the rates (in milligrams/day) were normalized to the net energy produced in each case. The production and destruction rates of Pu-241 in the MOX assembly are similar throughout irradiation, so there is only a slight decrease in the Pu-241 mass from beginning- (BOL) to end-of-life (EOL). The bulk of the Am-241 chain consumption in MOX fuel comes from transmutation of the Am-241.

Figure 15 shows that both the production and destruction rates of Pu-241 in the IMF assembly increase with fuel irradiation, although the production rate increases less rapidly. (The same behavior is exhibited in the MOX fuel, but it is not as pronounced.) Consequently, there is initially a net production of Pu-241 in the recycle assemblies, but with continued irradiation the ongoing destruction of Pu-241 will exceed its production. For the IMF assembly, this transition occurs relatively early in the irradiation history (at about 1/3 of the discharge burnup) so that a greater reduction of the Am-241 chain mass is realized in IMF relative to MOX.

The parameters controlling this behavior are illustrated in Figure 16. As Pu-240 is converted to Pu-241, the declining Pu-240/Pu-241 ratio in the fuel slows the Pu-241 production relative to its destruction. Also, the neutron spectrum becomes softer with

Table 7. Assembly-Averaged, One-Group Cross Sections at Beginning-of-Life.
Derived from WIMS8 (JEF 2.2 library) at 14.9 full-power days irradiation, 0 ppm soluble boron.

Nuclide	*UO_2 Assembly*			*MOX-1 Assembly (5-Year Cooled TRU)*			*MOX-1 Assembly (20-Year Cooled TRU)*			*IMF-1 Assembly (5-Year Cooled TRU)*			*IMF-1 Assembly (20-Year Cooled TRU)*		
	σ_{abs} (barns)	$\sigma_{fission}$ (barns)	σ_f/σ_a	σ_{abs} (barns)	$\sigma_{fission}$ (barns)	σ_f/σ_a	σ_{abs} (barns)	$\sigma_{fission}$ (barns)	σ_f/σ_a	σ_{abs} (barns)	$\sigma_{fission}$ (barns)	σ_f/σ_a	σ_{abs} (barns)	$\sigma_{fission}$ (barns)	σ_f/σ_a
Am241	**116.7**	**1.3**	**0.01**	**24.4**	**0.7**	**0.03**	**16.8**	**0.7**	**0.04**	**44.6**	**0.8**	**0.0**	**33.4**	**0.8**	**0.0**
Am242m	758.0	616.1	0.81	83.3	69.1	0.83	52.9	44.2	0.83	197.3	161.9	0.8	138.7	114.3	0.8
Am243	52.8	0.4	0.01	25.3	0.5	0.02	18.9	0.5	0.03	40.5	0.5	0.0	33.8	0.5	0.0
Cm242	5.3	1.1	0.21	4.1	0.9	0.22	3.8	0.9	0.24	4.8	1.0	0.2	4.6	0.9	0.2
Cm243	87.0	73.9	0.85	41.3	35.3	0.86	33.9	28.9	0.85	62.0	53.0	0.9	54.7	46.8	0.9
Cm244	18.1	1.0	0.05	12.7	1.0	0.08	10.8	1.0	0.09	17.5	1.0	0.1	16.1	1.0	0.1
Cm245	135.1	116.9	0.87	26.5	22.8	0.86	20.3	17.4	0.86	49.0	42.2	0.9	38.6	33.2	0.9
Np237	34.7	0.5	0.01	15.1	0.6	0.04	12.2	0.6	0.05	23.0	0.6	0.0	19.6	0.6	0.0
Np239	14.5	0.6	0.04	11.5	0.7	0.06	10.5	0.7	0.07	14.7	0.7	0.0	14.0	0.7	0.0
Pu238	31.0	2.4	0.08	7.2	1.8	0.25	6.0	1.8	0.30	11.7	1.9	0.2	9.5	1.9	0.2
Pu239	166.8	106.0	0.64	19.6	12.6	0.65	13.4	8.7	0.65	41.2	26.5	0.64	29.8	19.2	0.64
Pu240	234.2	0.6	0.00	15.0	0.7	0.05	9.6	0.7	0.07	30.9	0.7	0.0	22.4	0.7	0.0
Pu241	**145.3**	**108.5**	**0.75**	**25.4**	**19.5**	**0.77**	**19.8**	**15.3**	**0.77**	**47.3**	**35.9**	**0.8**	**37.5**	**28.6**	**0.8**
Pu242	30.1	0.4	0.01	9.0	0.5	0.06	6.6	0.5	0.08	15.6	0.5	0.0	12.6	0.5	0.0
U233	58.5	51.9	0.89	20.2	17.4	0.86	16.4	14.1	0.86	31.8	27.5	0.9	27.2	23.4	0.9
U234	20.2	0.5	0.03	13.0	0.6	0.05	11.4	0.6	0.05	18.6	0.6	0.0	17.2	0.6	0.0
U235	48.7	39.8	0.82	12.4	9.0	0.73	10.2	7.3	0.71	20.3	15.1	0.7	16.8	12.3	0.7
U236	8.6	0.3	0.04	5.3	0.3	0.06	4.8	0.3	0.07	10.2	0.4	0.0	9.6	0.4	0.0
U237	35.4	0.6	0.02	9.2	0.6	0.06	7.5	0.6	0.08	14.9	0.6	0.0	12.3	0.6	0.0
U238	1.0	0.1	0.10	0.9	0.1	0.14	0.9	0.1	0.14	8.5	0.1	0.0	8.2	0.1	0.0

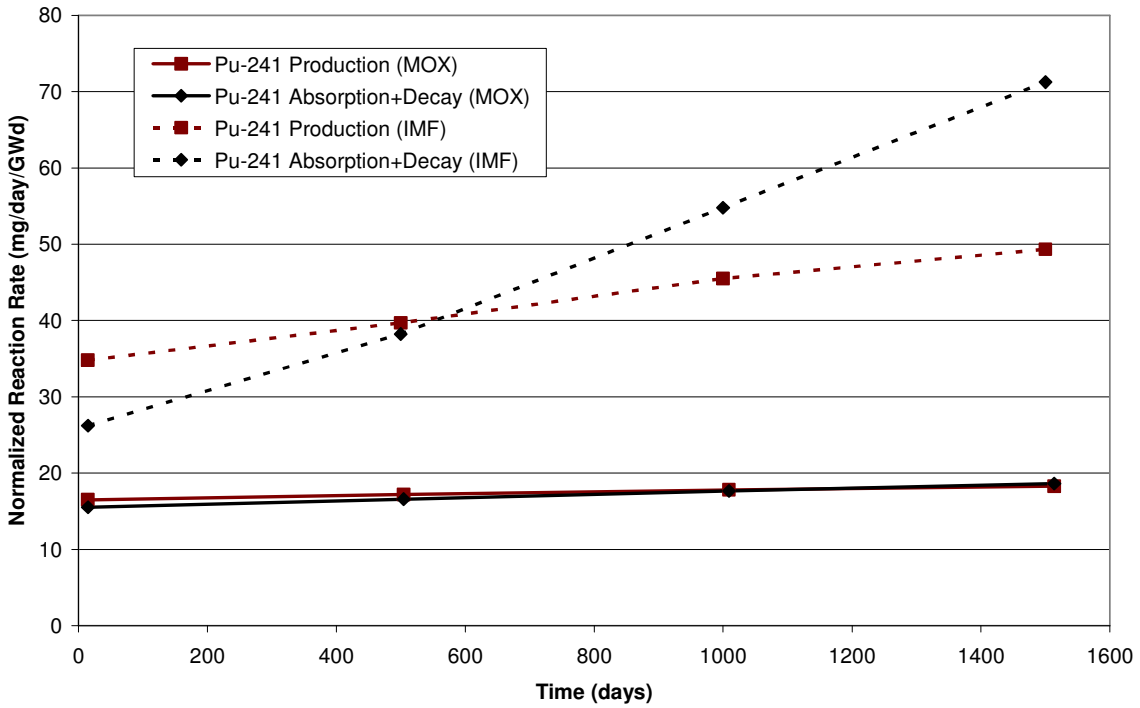

Figure 15. Pu-241 Production and Destruction Rates in First-Recycle MOX and IMF Assembly Fabricated with 5-Year Cooled TRU. Reaction rates have been normalized to the net energy produced by the fuel material before disposal.

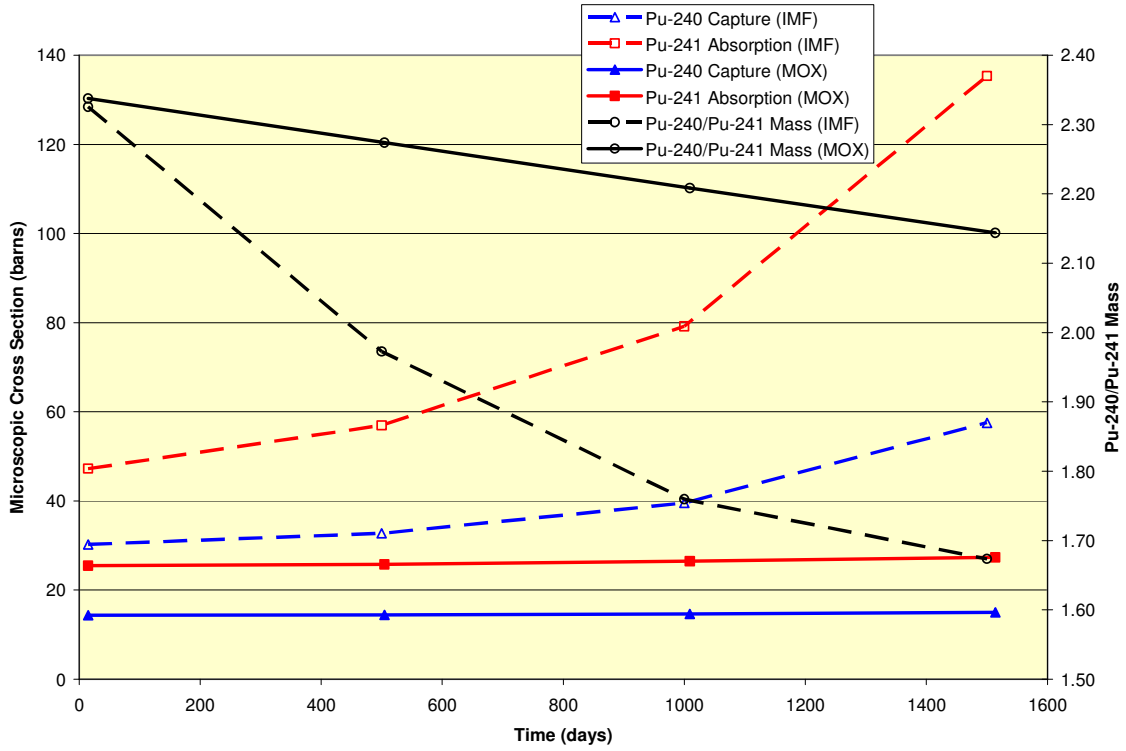

Figure 16. Pu-240 Capture and Pu-241 Absorption Cross Sections, and Pu-240/Pu-241 Mass Ratio in First Recycle MOX and IMF Assemblies.

irradiation of the TRU-bearing fuels due to the consumption of plutonium in the assembly; this effect is particularly severe in the case of IMF fuel, in which the fast-to-thermal flux declines from 7.6 at BOL to 2.7 at EOL. (Typically, the spectrum hardens with depletion in UO_2 due to the net production of plutonium.) The softening of the neutron spectrum increases the cross sections for all plutonium isotopes, but this effect is greater for Pu-241 absorption (and Am-241 absorption), which has a higher cross section at lower energies than Pu-240 capture; see Figure 17. Although the same trends exist in the MOX fuel, they are less pronounced because there is less net plutonium consumption (and spectrum softening) in the MOX fuel due to the ongoing plutonium production from U-238 conversion.

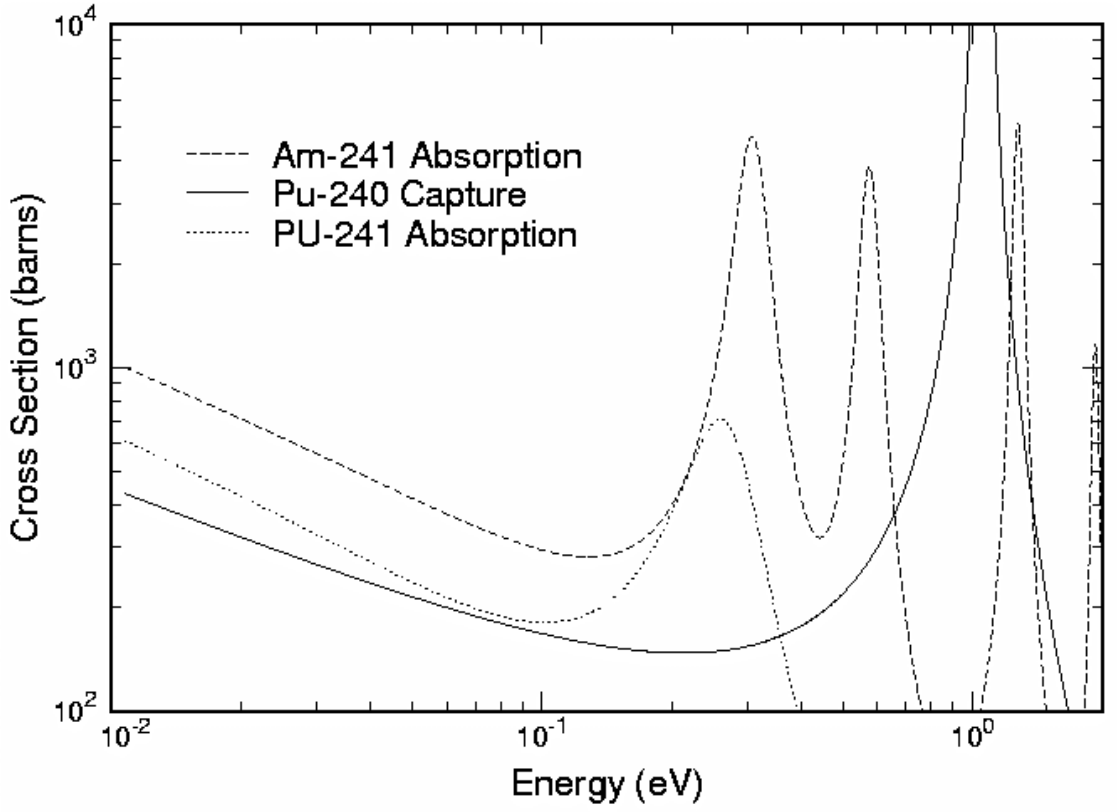

Figure 17. Energy Dependence of Pu-240, Pu-241, and Am-241 Cross Sections.

Transuranic recycling in the CORAIL strategy is conceptually similar to that using homogeneous MOX assemblies, in that the initial source of the recycled transuranics is spent UO_2 and the material is recycled in a MOX fuel form. Even so, the CORAIL strategy yields a greater repository benefit for a given number of recycles, and has the potential for achieving a greater benefit overall because more recycles can be practiced before recycling must be terminated due to reactor operations and safety considerations. In order to understand the physics behind the improved repository performance in the CORAIL strategy, the masses of the key contributors to the repository heating in the direct-disposed CORAIL and MOX assemblies after a single recycle of Pu+Np+Am are compared in Table 8. As discussed above, the masses have been normalized to the net energy extracted prior to disposal (kg/TWd).

Table 8. TRU Masses (kg/TWd) and TRU Mass Fraction in Direct-Disposed CORAIL and MOX Assemblies After Single Recycle of Pu+Np+Am. Masses are normalized to the net energy produced prior to disposal.

	TRU	Pu238		Pu239		Pu240		Am-241 Chain	
CORAIL Assembly									
UOX pins	93	2	(2.4%)	47	(50.4%)	20	(21.1%)	13	(14.0%)
MOX pins	100	7	(7.2%)	29	(29.2%)	24	(24.1%)	15	(15.1%)
CORAIL Total	**194**	**9**	(4.8%)	**76**	(39.4%)	**44**	(22.6%)	**28**	(14.6%)
MOX Assembly	**203**	**13**	(6.2%)	**77**	(37.8%)	**51**	(25.0%)	**31**	(15.2%)

The loading of first-recycle TRU in the pins of the homogeneous MOX assembly is 17.4 wt.%, compared with only 4.09 wt.% in the CORAIL assembly (51 GWd/MT case). Because the TRU loading in the CORAIL MOX pins is lower, the neutron spectrum is significantly softer than that in the homogeneous assembly MOX, increasing the Np-237 and Am-241 capture cross sections and yielding a higher fraction of Pu-238 in the spent CORAIL MOX. However, since only about half of the TRU sent to the repository comes from the MOX pins (the remainder comes from the spent UO_2 pins in the CORAIL assembly), the total normalized Pu-238 mass in the CORAIL assembly is ~25% lower than that in the discharged MOX assembly.

As discussed above, softening the neutron spectrum increases the destruction rates of Pu-241 and Am-241 relative to their production. At the same time, the reduced Pu-240 loading in the CORAIL MOX pins reduces the self-shielding in the large capture resonance near 1 eV, increasing the Pu-240 capture cross section. These effects effectively compensate for one another, so that the fraction of Am-241 chain nuclides in the CORAIL MOX pins is similar to that in the homogeneous MOX pins. However, blending the CORAIL MOX and UO_2 pins yields an 8% reduction in the normalized Am-241 chain mass relative to the homogeneous MOX assembly. Likewise, the normalized Pu-240 mass is 14% lower.

The lower discharged masses of Pu-238, Am-241, and Pu-240 in the CORAIL assembly relative to the homogeneous MOX improve the performance of this recycling approach for increasing the repository loading. The relevant differences between the MOX and CORAIL strategies which contribute to the improved repository performance are that the directly-disposed spent fuel at the end of the CORAIL recycling campaign consists of spent UO_2 and MOX fuel pins, and the CORAIL strategy (as formulated in this study) utilizes a lower TRU loading in the MOX pins. Of course, the improved benefit comes with the expense of higher UO_2 enrichments which exceed the current limit of 5 wt.% U-235 for U. S. fabrication facilities.

The TRU loading in the homogenous MOX pins was determined based on an assembly-level calculation to simplify the analysis (effectively, a full-core loading of MOX). In practice, the MOX assemblies would be loaded in "mixed" core (UO_2 and MOX fuel), which would reduce the TRU loading needed to meet the operating cycle requirements. Thus, the repository benefit for the homogeneous MOX strategy could be

improved, with the CORAIL results representing an upper-bound on the repository benefit derived from MOX recycling.

IV.2. Impact of Cooling Time Before Spent Fuel Reprocessing and Recycling

After five years, the spent fuel has cooled sufficiently to allow aqueous reprocessing (e.g., UREX+), but longer cooling times might be preferred in practice to balance reprocessing and fabrication mass flows. In order to assess the impact of longer cooling intervals on the repository benefit, calculations have also been performed assuming 20 years post-irradiation cooling before spent UO_2 reprocessing and TRU recycling.

Since Am-241 is the primary contributor to the decay heat of disposed reactor-grade TRU in the time-frame of interest for repository heating (100-1500 years), LWR transmutation strategies target the destruction of Pu-241 and Am-241 to reduce the amount of Am-241 chain nuclides disposed in the repository waste. However, while Pu-241 is primarily destroyed by fission, ~75% of neutron absorptions in Am-241 yield Pu-238 (see Figure 4) which, while not as problematic as the Am-241, still contributes to the repository heating. Since the ongoing decay of Pu-241 prior to reprocessing (or following fabrication of fresh MOX or IMF) increases the relative fraction of Am-241 in the recycled TRU (see Table 4), it appears preferable from the perspective of reducing the repository heating to recycle the Pu-241 back into the reactor as soon as practical. Furthermore, a higher initial loading of longer-cooled TRU will be needed to maintain the same reactivity-limited assembly discharge burnup because Am-241 acts as a neutron poison in an LWR spectrum ($\sigma_f/\sigma_a \sim 0.03$). This will increase the heat and radiation sources of the fabricated fuel when recycling is delayed, complicating shielding requirements and fuel handling procedures.

Table 9 provides charge and discharge (after 5-years post-irradiation cooling) actinide masses for UO_2, and first-recycle MOX and IMF assemblies. The key parameter for comparison between cases is the discharge mass normalized to the net energy produced (kg/TWd), since this reflects the amount of each nuclide disposed in the repository while taking into account the displacement of directly-disposed UO_2 in the recycle scenarios. For the discharged MOX fuel, the normalized Am-241 chain mass is not affected by the longer cooling time; similar behavior was also observed for the CORAIL case. The higher concentration of Am-241 in the repository waste is balanced by a reduction of the normalized Pu-241 mass. There are, however, slight increases in the normalized discharge masses of Pu-238, Pu-239, and Pu-240 when the cooling time is increased, primarily due to the higher TRU loading in the fabricated fuel. Consequently, increasing the cooling time before reprocessing reduces the repository loading benefit due to a single recycle from 1.09 to 1.04 for MOX, and from 1.22 to 1.16 for CORAIL fuel. Thus, there is a small penalty on the repository loading benefit associated with recycling older transuranic material in a MOX fuel form.

After a single recycle in IMF, the normalized Am-241 chain mass discharged to the repository is 65% higher when reprocessing/recycling is delayed from 5 to 20 years after UO_2 discharge, and the Pu-238 mass sent to the repository is 50% higher. The impact of the increased discharge masses of these nuclides on the disposed waste decay heat can be clearly seen in Figure 14. Likewise, the normalized Pu-239 and Pu-240 masses are 350% and 60% greater, respectively. Relative to the reference UO_2 case, the repository

Table 9. UO_2, First Recycle MOX, and First Recycle IMF Assembly Masses.

	UO_2 4.3 wt.% U-235	*First Recycle MOX 17.4 wt.% Pu+Np+Am/HM (5-Year Cooled TRU)*			*First Recycle MOX 27.7 wt.% Pu+Np+Am/HM (20-Year Cooled TRU)*			*First Recycle IMF 21.3 wt.% Pu+Np+Am in ZrO_2 matrix (5-Year Cooled TRU)*			*First Recycle IMF 30.0 wt.% Pu+Np+Am in ZrO_2 matrix (20-Year Cooled TRU)*		
Nuclide	**Dischg. (kg/TWd)**	**Charge[a] (kg)**	**Dischg.[b] (kg)**	**Dischg. [c] (kg/TWd)**	**Charge (kg)**	**Dischg. (kg)**	**Dischg. (kg/TWd)**	**Charge (kg)**	**Dischg. (kg)**	**Dischg. (kg/TWd)**	**Charge (kg)**	**Dischg. (kg)**	**Dischg. (kg/TWd)**
Am241	9	3.52	4.22	12	11.98	9.60	18	1.85	0.92	5	5.87	2.90	11
Am242m	0	0.01	0.07	0	0.01	0.23	0	0.00	0.01	0	0.00	0.04	0
Am243	4	1.22	1.62	5	1.94	2.38	4	0.64	1.14	6	0.95	1.45	5
Americium	**12**	**4.74**	**5.92**	**17**	**13.93**	**12.21**	**23**	**2.50**	**2.06**	**11**	**6.83**	**4.39**	**16**
Cm243	0	0.00	0.01	0	0.00	0.03	0	0.00	0.01	0	0.00	0.03	0
Cm244	1	0.000	0.776	2	0.000	0.884	2	0.000	0.904	5	0.000	0.960	4
Cm245	0	0.00	0.14	0	0.00	0.14	0	0.00	0.13	1	0.00	0.19	1
Curium	**1.5**	**0.00**	**0.93**	**2.7**	**0.00**	**1.05**	**2.0**	**0.00**	**1.04**	**5.5**	**0.00**	**1.17**	**4.3**
Neptunium	**12**	**3.84**	**2.60**	**8**	**6.12**	**4.32**	**8**	**2.02**	**0.66**	**3**	**3.00**	**1.33**	**5**
Pu238	6	1.88	4.26	13	2.66	7.20	14	0.99	2.33	12	1.30	4.82	18
Pu239	121	38.49	26.19	77	61.30	45.64	86	20.27	1.27	7	30.06	6.89	25
Pu240	57	18.21	17.30	51	29.26	28.74	54	9.59	4.80	25	14.35	10.89	40
Pu241	27	7.78	6.29	18	6.03	6.64	13	4.10	2.17	11	2.96	4.31	16
Pu242	17	5.34	5.75	17	8.50	8.61	16	2.81	4.23	22	4.17	4.96	18
Plutonium	**228**	**71.70**	**59.79**	**175**	**107.75**	**96.83**	**183**	**37.76**	**14.81**	**78**	**52.84**	**31.87**	**117**
U235	151	3.13	2.06	6	2.74	1.99	4	0.00	0.02	0	0.00	0.03	0
U238	18,059	375.44	363.56	1,066	328.60	318.51	602	0.00	0.00	0	0.00	0.00	0
Uranium	**18,325**	**381.01**	**368.20**	**1,080**	**333.49**	**322.97**	**610**	**0.02**	**0.16**	**1**	**0.03**	**0.32**	**1**
Am-241 Chain	**36**	**11.30**	**10.51**	**31**	**18.01**	**16.25**	**31**	**5.95**	**3.09**	**16**	**8.83**	**7.21**	**27**
TRU	**255**	**80.29**	**69.23**	**203**	**127.81**	**114.41**	**216**	**42.28**	**18.57**	**97**	**62.67**	**38.76**	**143**
HM		**461.30**	**437.42**		**461.30**	**437.39**		**42.30**	**18.74**		**62.70**	**39.08**	
Am-241/TRU		***4.4%***			***9.4%***			***4.4%***			***9.4%***		

[a]Assembly charged to the reactor two years after spent UO_2 reprocessing and assembly fabrication..
[b]Assembly discharge values reported after five years post-irradiation cooling.
[c]Normalized to net energy extracted from TRU. Net burnup is: UO_2, 51 GWd/MT; MOX, 739 GWd/MT; MOX(20-yr cooled), 1148 GWd/MT; IMF, 4507 GWd/MT; IMF(20-yr cooled), 4331 GWd/MT.

capacity is only 19% higher for the longer-cooled case, compared with a 79% loading benefit for a single recycle of 5-year cooled TRU in IMF. Thus, it is clear that *delaying spent fuel reprocessing and recycling significantly reduces the benefits of a single IMF recycle*.

The penalty associated with delaying TRU recycling in IMF is largely due to the increased Am-241 chain mass disposed in repository waste. About half of this increase is due to an increase of the normalized Am-241 mass at discharge, and the remainder is due to an increase in the discharged Pu-241 mass for the longer-cooled IMF case. This latter increase is counter-intuitive since the normalized mass at charge is lower for the longer-cooled case; also, the opposite behavior is exhibited for the MOX fuel (the discharge Pu-241 mass is lower when the cooling time is extended). This behavior is worth examining.

Figure 18 compares the Pu-241 production and destruction rates in IMF assemblies fabricated with 5- and 20-year cooled TRU. Relative to the 5-year cooled case, the normalized Pu-241 destruction rate in the 20-year cooled case is reduced by roughly 50% over the irradiation history of the IMF assembly due to a number of effects. First, there is a lower Pu-241 loading in the assembly at reactor charge. Also, the higher TRU loading in the assembly yields a harder neutron spectrum (and increases neutron self-shielding for certain nuclides), which lowers the actinide cross sections as shown in Table 7. As fissile Pu-239 is consumed and the inventory of fission products increases over the irradiation history, the neutron flux must be increased to maintain the assembly power. The Pu-239 consumption is nearly 95% in the 5-year cooled case, but is only about 75% for the 20-year cooled case; a higher initial loading of fissile Pu-239 is needed to

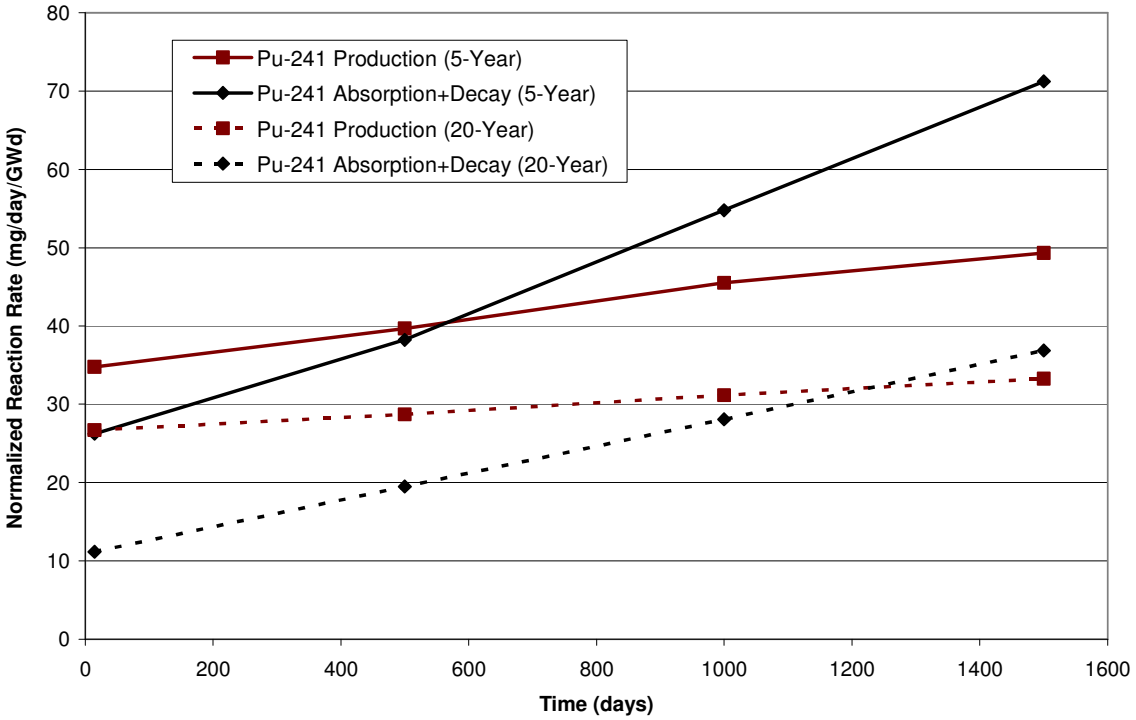

Figure 18. Pu-241 Production and Destruction Rates in First-Recycle IMF Assembly Fabricated with 5- and 20-Year Cooled TRU. Reaction rates have been normalized to the net energy produced by the fuel material before disposal.

compensate for the increased parasitic absorption in Am-241. Consequently, the neutron flux required to maintain the same power level is lower in the longer-cooled case, which further reduces the Pu-241 destruction rate. The production rate of Pu-241 is also lower due to the reduction of the cross sections and flux, but is reduced by less than 30% relative to the 5-year cooled case. Thus, the balance between Pu-241 production and destruction occurs much later in the irradiation, resulting in a 80% increase in the normalized Pu-241 mass from beginning- to end-of-life.

IV.3. Impact of Transuranic Separations Strategy

While the present study has focused on plutonium, neptunium, and americium recycling in LWRs for management of transuranics in CSNF, previous studies have considered the recycling of plutonium only, plutonium+neptunium, and all transuranic species in MOX fuel. The disposed masses of transuranics *after a single recycle* are cross-compared in Table 10 for a variety of separations strategies. To facilitate the comparison, the masses have been normalized to the net energy produced by the transuranics and their source UO_2 (kg/TWd). The masses include all nuclides in the spent recycled assembly, as well as any nuclides in a stream which "bypasses" the LWR transmuter following spent UO_2 reprocessing and transuranic separation.

The results for all cases were derived from assembly-level calculations in WIMS8 which simulate a full-core loading of the specified fuel form, except for the case with plutonium recycling labeled "MOX/UO_2." In that case, the discharge masses were derived for a MOX assembly irradiated in a mixed lattice of one MOX and 3 UO_2 assemblies. While the actinide masses in the spent MOX assembly will vary with the initial transuranic loading and assembly burnup, there is good agreement between these two sets of results because the masses have been normalized to the net energy produced, which takes these factors into account. Larger differences do appear for Pu-241, Am-241, and Cm, but these differences are simply due to the longer cooling time before tabulation of the results in the mixed-lattice depletion analysis. The agreement observed here confirms that simple assembly-level calculations can provide good predictions of the normalized discharged actinide masses from transuranic-bearing fuels, regardless of the particular details of the core arrangement (i.e., full-core or partial-core loadings of MOX fuel).

All cases provide reductions in the normalized TRU and Pu-239 masses, ranging from 30% Pu-239 consumed for MOX fabricated with all transuranics to 97% consumption for the inert-matrix fuel form with Pu+Np recycling. There is also a reduction in the disposed plutonium mass, which provides the benefit of lowering the attractiveness of the repository as a "plutonium mine." On the other hand, all cases show an increase in the normalized curium mass relative to the once-through fuel cycle, even when curium is recycled in the MOX fuel. The increase in curium mass is larger when americium is homogeneously recycled. Since Am-241 acts as a neutron poison, a higher plutonium loading is required in the fabricated fuel to meet the same reactor cycle length. Consequently, there is a higher initial loading of Pu-242, which serves as the "gateway" to Cm-244 in the transmutation chains. In any event, the consequence of increasing the disposal of curium in the repository is outweighed by the benefits of reducing the amounts of Am-241 and Np-237 in the facility.

Table 10. Comparison of Disposed Transuranic Masses Normalized to Net Energy Produced for Various LWR Recycle Strategies.

Fuel Form	**UO_2**	**MOX**	**MOX/UO_2**	**MOX**	**IMF**	**MOX**	**CORAIL**	**IMF**	**MOX**
Recycled TRU	N/A[1]	Pu[2]	Pu[2]	Pu,Np[1]	Pu,Np[1]	Pu,Np,Am[1]	Pu,Np,Am[1]	Pu,Np,Am[1]	Pu,Np, Am,Cm[2]
Cooling before UO_2 reprocessing	N/A	10 Years	10 Years	5 Years	5 Years	5 Years	5 Years	5 Years	10 Years
TRU/HM in Fresh Fuel	N/A	8.85%	8.00%	13.10%	100%	17.40%	1.30%[3]	100%	18.25%
Cooling before disposal	5 Years	5 Years	10 Years	5 Years	5 Years	5 Years	5 Years	5 Years	5 Years
Disposed Mass[4] (kg/TWd)									
Np237	12.1	18.6	18.8	7.4	3.0	7.6	7.4	3.5	11.4
Pu238	6.0	6.2	6.4	10.2	7.9	12.5	9.3	12.3	16.1
Pu239	121.3	60.2	62.0	68.9	3.0	76.8	76.0	6.7	85.2
Pu240	57.4	51.1	52.6	48.4	18.3	51.2	43.9	25.7	58.4
Pu241	27.0	20.5	15.8	19.4	7.6	18.5	20.7	11.4	18.5
Pu242	16.8	15.4	15.5	16.6	23.1	16.9	19.0	22.2	14.6
Am241	8.6	19.1	23.7	16.6	10.1	12.4	7.4	4.8	12.8
Cm	1.5	3.0	2.2	2.5	5.1	3.6	4.4	6.3	3.6
Am-241 Chain	*35.6*	*39.6*	*39.5*	*35.9*	*17.7*	*30.8*	*28.1*	*16.2*	*31.3*
Np-237 Chain	*47.7*	*58.2*	*58.3*	*43.4*	*20.7*	*38.5*	*35.5*	*19.7*	*42.7*
TRU	*255*	*202*	*204*	*197*	*87*	*205*	*193*	*99*	*225*

[1]Transuranic vector and TRU weight fraction in spent UO_2 derived from WIMS8 for 4.3 wt.% U-235 discharged at 51 GWd/MT and cooled 5 years.
[2]Transuranic vector and TRU weight fraction in spent UO_2 derived from ORIGEN2 (*pwrue* library) for 4.2 wt.% U-235 discharged at 50 GWd/MT and cooled 10 years.
[3]Average TRU loading in CORAIL assembly (MOX and UO_2 pins). TRU loading in MOX pins is 4.09 wt.%.
[4]Includes all transuranics discharged in spent fuel plus any "bypass" stream of actinides which are not recycled in the LWR.

When plutonium or plutonium+neptunium are recycled in MOX, the normalized mass of the Am-241 chain (Pu-241+Am-241) is increased relative to the once-through fuel cycle. This increase is particularly large for the plutonium recycling cases, and is attributed to the production of Pu-241 from Pu-240 conversion during MOX irradiation. The implication is that a plutonium mono-recycling campaign worsens the problem of nuclear waste disposal if the goal is reducing the heat load on the repository.

The largest benefit from a single recycle of transuranics is gained when americium is recycled along with the plutonium (and possibly the neptunium). Reductions in the disposed masses of the Am-241 chain (reduces repository heating) and Np-237 chain (reduces released dose) are realized from a single recycle, although these gains are considered marginal in the light of a continued utilization of nuclear power and generation of spent fuel.

V. CONCLUSIONS

The option of transuranic recycling in U. S. LWRs has the advantage of exploiting a large capacity of nuclear reactor systems and making progress in the near-term by demonstrating the benefits of a nuclear fuel cycle which incorporates spent fuel reprocessing and recycling. Experience has already been gained in European (particularly, France) and Japanese nuclear programs from utilizing MOX fuel fabricated with reactor-grade plutonium, separated from spent uranium and the other transuranics using the PUREX process. While employing this approach recovers valuable energy (from plutonium fission) which would be discarded in a once-through UO_2 fuel cycle, the issue of Am-241 disposition, which contributes to repository heating and essentially limits the repository loading, would remain.

In the present study, the effectiveness of limited homogeneous recycling of plutonium, neptunium, and americium in LWRs for improving the repository performance relative to a once-through fuel cycle has been investigated. The recycled transuranics were loaded into fuel pins in MOX, CORAIL (heterogeneous MOX and UO_2), and inert-matrix (IMF) fuel assemblies. Studies of the viability of using heterogeneous targets for americium and curium transmutation are also being performed within the AFCI program. Charge and discharge isotopic vectors and mass flows were determined, and the consequent impacts on safety parameters and repository heat load were estimated.

The basic assumptions employed in this study included:

- High-burnup UO_2 (51 or 45 GWd/MTHM)
- Minimal post-irradiation cooling time (5 years) before spent fuel reprocessing and TRU recycling
- Separated cesium, strontium, and depleted uranium sent to interim storage
- Separated curium assumed sent to repository for permanent disposal
- Direct disposal of spent fuel assemblies from the final recycle stage

For the MOX and IMF recycling strategies, the Pu+Np+Am in the charged assembly in recycle N was derived solely from discharged assemblies in recycle N-1. Both strategies exhibited a rather rapid burndown of the fissile plutonium. Consequently, the transuranic loading needed to maintain the reactor cycle length in successive recycles had to be increased beyond what is practical from the viewpoint of safety coefficients fuel handling parameters, but this approach is the most straightforward and predicts the maximum heat load benefit achievable for a given number of recycles. For the IMF strategy in particular, nothing more than a single recycle is considered feasible unless the recycled fuel is supported with reactor-grade material (i.e., that derived from spent UO_2) or low-enriched uranium. Future studies which refine the recycling strategies to include blending schemes or heterogeneous IMF/UO_2 assembly designs should be considered.

Because of the heterogeneous design of the CORAIL assembly, it is most practical to co-process the spent UO_2 and MOX pins; distinguishing and segregating spent UO_2 and MOX pins would be difficult. Thus, the CORAIL assembly concept incorporates a recycling strategy which blends reactor-grade and recycled TRU. Utilizing this approach, the material can be multi-recycled without encountering operations constraints. As presently formulated this approach slowly stabilizes the inventory of transuranics, but does not burn down the TRU in the existing stockpile.

Pu+Np+Am recycling in homogeneous MOX assemblies provides the smallest repository benefit for a fixed number of recycles, although this recycling strategy is the most straightforward approach given current experience in Europe and Japan. A single recycle in this fuel form yields a 9% increase in the effective loading of spent fuel in the repository based on thermal limits in the current "high-temperature operating mode." Continued recycling through up to 5 recycles of the Pu+Np+Am was considered, resulting in a repository loading increase of 49% relative to the once-through fuel cycle. However, the large transuranic loading at this point (>50% TRU/HM) presents challenges for fuel fabrication (materials), reactor operations (safety coefficients), and fuel handling (dose to workers).

The CORAIL concept provides a modest gain on the repository loading relative to the homogeneous MOX approach. If only a single recycle is practiced, the repository capacity can be theoretically increased by 20%, while if 5 recycles are practiced, there is about a 70% improvement relative to the once-through fuel cycle. The utilization of an inert-matrix fuel form such as solid solution $(TRU,Zr)O_2$ yields the greatest benefit because no additional transuranics are produced during the fuel irradiation. A single recycle provides a loading increase of 80%, but further recycles, although theoretically possible, are not considered practical without enhancing the recycling strategy.

Recycling plutonium and americium in MOX, CORAIL, and IMF fuel forms contributes directly to the repository loading benefit by reducing the masses of Am-241 chain nuclides (Pu-241+Am-241) disposed in nuclear waste, and their associated decay heat, relative to the once-through fuel cycle. Neptunium was also recycled in these scenarios because of the potential reduction of the released dose from the repository, although the dose reduction was not quantified in this study. However, recycling the neptunium penalizes the loading benefit somewhat due to the increased disposal of Pu-238 and its associated decay heat. An assessment of the trade-offs between dose reduction and increased decay heat due to neptunium recycling could be performed. The curium extracted from spent UO_2 was not recycled in the LWR recycling strategies evaluated here because of its added complications to fuel handling.

It was found that the longer reprocessing and recycling of the material in the spent UO_2 is delayed, the less is the achievable benefit. The consequences of delaying recycling are most severe for the IMF fuel form. Ongoing decay of fissile Pu-241 to Am-241 increases the required TRU loading, hardening the neutron spectrum in a way that negatively impacts the Pu-241 and Am-241 transmutation rates. Extending the cooling time from 5 to 20 years from reactor discharge to recycling of the Pu+Np+Am in IMF, the repository loading benefit is reduced by a factor of 4, or a 20% loading increase relative to a once-through fuel cycle. The current stockpile of spent nuclear fuel in the U. S. has an average age of about 15 years, based on a mass-weighting of spent fuel discharges beginning in 1968. Thus, the strategies investigated here for homogeneously recycling plutonium, neptunium, and americium in LWRs seem best suited for the youngest (most recently discharged) spent fuel.

Lastly, the impact of the transuranic separations strategy employed for actinide management was investigated by comparing the results obtained here with those obtained from earlier studies. In particular, the discharged actinide masses (normalized to the net energy produced by the transuranics and their source UO_2) after a single LWR recycle

were compared for Pu, Pu+Np, Pu+Np+Am, and all TRU separations strategies. Mono-recycling of separated plutonium burns fissile Pu-239 and would lower the attractiveness of the repository as a "plutonium mine." However, the normalized mass of Am-241 and Np-237 chain nuclides are actually increased by this approach relative to a once-through fuel cycle. The implication is that this elevates the long-term decay heat which limits the waste loading in the current repository design.

Recycling the americium appears necessary for an actinide management strategy that benefits the repository in terms of reduced heat load and reduced released dose, although the gains which can be realized from limited recycling in an LWR are considered marginal in light of a continued utilization of nuclear power and generation of spent fuel. Whether nuclear power grows or even continues at its current capacity, the gains realized here are not large enough to stave off the need for a second repository unless further recycling in LWRs or future, advanced systems is practiced. Nonetheless, actinide management in LWRs is sensible, particularly as a demonstration of spent fuel reprocessing and recycling technologies to be employed in Generation-IV systems and/or as a technique for reducing the hazardous components in legacy spent nuclear fuel.

However, challenges to transuranic recycling in current-generation LWR systems do exist and these must be considered either as topics for future technical work or as factors in making decisions regarding the viability of employing LWRs for actinide management. There is, first of all, the issue of fuels development and utilizing fuel forms which have not been commercially fabricated or extensively tested. MOX fuel is being used in international nuclear programs, but this has been limited to separated plutonium; inert-matrix fuels are just now being considered, and questions about reprocessing on the back-end of the fuel cycle must be addressed. Second, there are the concerns over the proliferation-resistance of utilizing a fuel cycle based on separation of transuranics from spent nuclear fuel. There does not appear to be a single technical argument to address all concerns, but, rather, intrinsic (e.g., isotopics) and extrinsic (e.g., security) safeguards will be necessary. Third, there remains the fundamental question of whether U. S. nuclear utilities will consider recycled fuel to be an attractive resource. At the heart of this issue are reliability (how will the types of fuels considered affect day-to-day reactor operations), reactor safety (are current safety systems designed for UO_2 fuel adequate), and economics (what will be the ultimate cost or benefit to the utility).

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